PB84-118504

International Meeting on Radon-Radon Progeny Measurements - Proceedings Held on August 27-28, 1981

Dingle Associates, Inc., Washington, DC

Prepared for

Environmental Protection Agency, Washington, DC

Sep 83

U.S. Department of Commerce
National Technical Information Service

NTIS

TECHNICAL REPORT DATA
(Please read Instructions on the reverse before completing)

1. REPORT NO.
520/5-83-021

2.

3. RECIPIENT'S ACCESSION NO.
PB84 118504

4. TITLE AND SUBTITLE
International Meeting on Radon-Radon Progeny Measurements Proceedings

5. REPORT DATE

6. PERFORMING ORGANIZATION CODE

7. AUTHOR(S)

8. PERFORMING ORGANIZATION REPORT NO.

9. PERFORMING ORGANIZATION NAME AND ADDRESS
Samoas box 12

10. PROGRAM ELEMENT NO.

11. CONTRACT/GRANT NO.

12. SPONSORING AGENCY NAME AND ADDRESS
U.S. Environmental Protection Agency
401 M Street, S.W.
Washington, D.C. 20460

13. TYPE OF REPORT AND PERIOD COVERED

14. SPONSORING AGENCY CODE

15. SUPPLEMENTARY NOTES

16. ABSTRACT

With the emergence of worldwide interest in radon-radon progeny levels in structures, many different methods and instruments for determining these levels have been developed. The Office of Radiation Programs (ORP), of the U.S. Environmental Protection Agency (EPA), has been developing a program to determine the significance of exposure from this radiation source. It has been ORP's objective to use appropriate measurement methods to assure that the results of studies are meaningful and to interpret the results of previous studies so that the limitations of the measurement methods are understood. With this objective in mind, EPA sponsored a two-day international meeting on radon and radon progeny measurements. The purpose of the meeting was to promote and interchange of information between international experts. The meeting format included formal presentations and directe discussions. Eighteen papers were presented on such topics as measurement methods and instrumentation and surveys and measurement programs using such instruments and methods. This publication contains the complete texts of 15 papers, abstracts of two papers, and edited transcripts of the two open discussions: "Instrumentation and Measurement Methods" and "Measurements and Related Topics."

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group

18. DISTRIBUTION STATEMENT
Unlimited

19. SECURITY CLASS *(This Report)*
unclassified

20. SECURITY CLASS *(This page)*
unclassified

21. NO. OF PAGES
278

22. PRICE

EPA Form 2220-1 (Rev. 4-77) PREVIOUS EDITION IS OBSOLETE

PB84-118504

United States
Environmental Protection
Agency

Office of
Radiation Programs
Washington, DC 20460

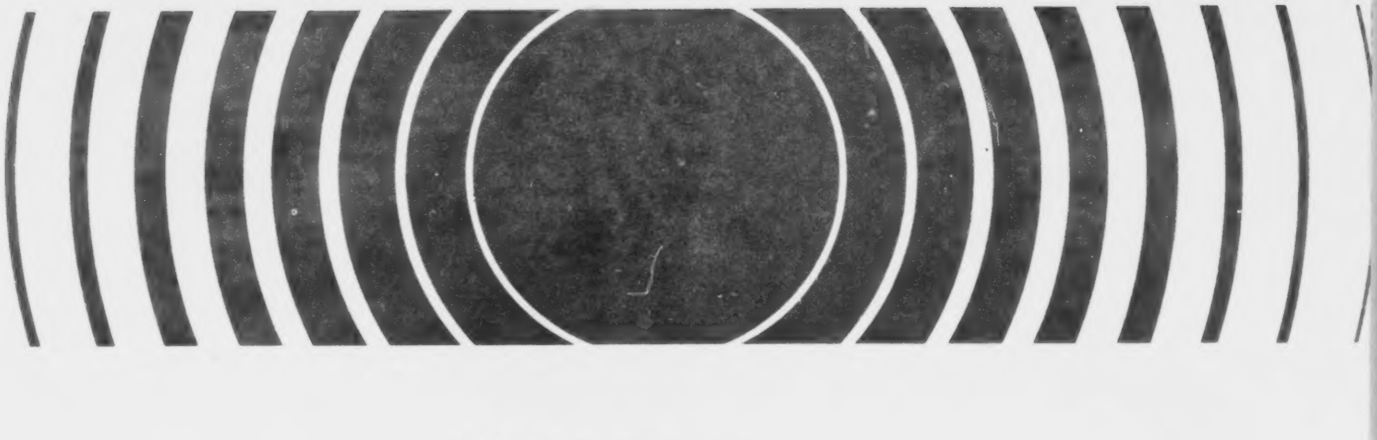
EPA 520/5-83/021
September 1983

Radiation

EPA

International Meeting on Radon-Radon Progeny Measurements

Proceedings



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NATIONAL TECHNICAL
INFORMATION SERVICE
U.S. DEPARTMENT OF COMMERCE
SPRINGFIELD, VA. 22161

INTERNATIONAL MEETING ON RADON-RADON PROGENY MEASUREMENTS
PROCEEDINGS

August 27-28, 1981

Sponsored By

U.S. Environmental Protection Agency
Office of Radiation Programs
Washington, D.C.

Prepared By

Dingle Associates, Inc.
1625 Eye Street, N.W., Suite 915
Washington, D.C. 20006

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INTRODUCTION

With the emergence of worldwide interest in radon-radon progeny levels in structures, many different methods and instruments for determining these levels have been developed. The Office of Radiation Programs (ORP), of the U.S. Environmental Protection Agency (EPA), has been developing a program to determine the significance of exposure from this radiation source. It has been ORP's objective to use appropriate measurement methods to assure that the results of studies are meaningful and to interpret the results of previous studies so that the limitations of the measurement methods are understood.

With this objective in mind, EPA sponsored a two-day international meeting on radon and radon progeny measurements. The purpose of the meeting was to promote an interchange of information between international experts. The meeting was held August 27-28, 1981, in Montgomery, Alabama, at the Holiday Inn - State Capitol. Mr. Charles R. Phillips, Chief, Technical Support Branch of the Eastern Environmental Radiation Facility (EERF) was the meeting coordinator.

In addition to the 48 participants who represented Federal and State government offices (both U.S. and abroad) and private industry, some 15 attendees included personnel from the Eastern Environmental Radiation Facility (the host site). The countries of Australia, Canada, Federal Republic of Germany, Great Britain, and Sweden were represented.

The meeting format included formal presentations and directed discussions. Eighteen papers were presented on such topics as measurement methods and instrumentation and surveys and measurement programs using such instruments and methods. Participants also toured the EERF on the last afternoon of the meeting. EERF personnel were on hand to host the tour and answer questions from the group.

An agenda and list of participants are included in the succeeding pages. The proceedings that follow include the complete texts of 15 papers, abstracts of two papers, and edited transcripts of the two open discussions: "Instrumentation and Measurement Methods" and "Measurements and Related Topics."

U.S. Environmental Protection Agency

INTERNATIONAL MEETING ON RADON-RADON PROGENY MEASUREMENTS

August 27-28, 1981

Holiday Inn - State Capitol
Montgomery, Alabama

AGENDA

WEDNESDAY, August 26, 1981	
6:00 - 8:00 p.m.	REGISTRATION
THURSDAY, August 27, 1981	
8:00 - 9:00 a.m.	REGISTRATION
9:00 - 9:20 a.m.	WELCOME and OPENING Introduction of Alabama Officials and Welcome from the State of Alabama Charles R. Phillips, Meeting Coordinator Opening Remarks David M. Rosenbaum, Ph.D, Office Director Office of Radiation Programs U.S. Environmental Protection Agency
9:20 - 12:25 p.m.	INSTRUMENTATION AND MEASUREMENT METHODS: Session I Chair: Peter G. Groer
9:20 - 10:00 a.m.	"Environmental Measurements Laboratory Developed Instrumentation"* *by Andreas George*
10:00 - 10:25 a.m.	"The EWLM II, A Measurement and Data Acquisition System for Radon-Daughters" *by Donald J. Keefe*
10:25 - 10:45 a.m.	Break
10:45 - 11:10 a.m.	"The Measurement of Radon-222 in Air by Direct Extraction into a Liquid Scintillator" *by Howard M. Prichard*

*Invited paper

11:10 - 11:35 a.m.	"Application of Surface Barrier Detectors for the Measurement of Environmental Radon and Radon Daughters in Air" *by Andreas Wicke*
11:35 - 12:00 noon	"Recent Advances in the Design and Use of Passive Radon Monitors" *by Lyle A. Rathbun*
12:00 - 12:25 p.m.	"Track Etch Detector Calibrations and Field Results" *by James E. Gingrich*
12:25 - 1:45 p.m.	Lunch
1:45 - 3:00 p.m.	INSTRUMENTATION AND MEASUREMENT METHODS: Session II Chair: Ron Collé
1:45 - 2:10 p.m.	"Assessment of Radon Measurement Techniques Used for Uranium Exploration" *by John C. Pacer*
2:10 - 2:35 p.m.	"A Review of Some Exact Methods for Rn-222 Daughter Measurement" *by Peter G. Groer*
2:35 - 3:00 p.m.	"Track Etch Calibration Design and Results of a Pilot Study"* *by Susan Hinkins*
3:00 - 3:20 p.m.	Break
3:20 - 5:20 p.m.	Open Discussion: INSTRUMENTATION AND MEASUREMENT METHODS Moderator: Peter G. Groer Panel: Speakers for Sessions I and II
7:00 - 11:00 p.m.	Riverboat Cruise and Dinner

FRIDAY, August 28, 1981

8:30 - 10:55 a.m.	RADON AND RADON PROGENY MEASUREMENTS Chair: Andreas George
8:30 - 8:55 a.m.	"Measuring Radon Source Magnitude in Residential Buildings" *by William W. Nazaroff*

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*Invited paper

8:55 - 9:20 a.m.	"Field and Laboratory Measurements of Radon Concentrations in Maine Houses" *by Charles T. Hess*
9:20 - 9:45 a.m.	"Applications of Single Grab Sample Measurements to the Decision-Making Process Involved in Choosing Houses for Remedial Action"* *by Arthur Scott*
9:45 - 10:05 a.m.	Break
10:05 - 10:30 a.m.	"Airborne Rn-222 in Buildings Constructed with High Radium-Content Concrete Blocks" *by Bernd Kahn*
10:30 - 10:55 a.m.	"Environmental Radon Investigations in Sweden" *by Gustav Akerblom*
10:55 - 1:00 p.m.	RELATED TOPICS Chair: Bernd Kahn
10:55 - 11:20 a.m.	"A Simple Procedure for Calibration of Continuous Flow-Through Radon Monitors" ** *by Erling Stranden*
11:20 - 11:45 a.m.	"Effect of Air Circulation and Dust Removal on Indoor Radon Decay Product Concentrations" *by Melvin W. First*
11:45 - 12:10 p.m.	"Some Measurements of the Equilibrium Factors in Houses" *by Richard E. Toohey*
12:10 - 12:35 p.m.	"Regional Environmental Documentation of Natural Radiation in Sweden" *by Carole Wilson*
12:35 - 1:00 p.m.	"An Apparatus for Calibrating Passive Integrating Radon Monitors" *by Robert A. Washington*
1:00 - 2:00 p.m.	Lunch
2:00 - 4:00 p.m.	OPEN DISCUSSION: MEASUREMENTS AND RELATED TOPICS Moderator: Arthur Scott Panel: Speakers for Measurements and Related Topics
4:00 p.m.	Closing

*Invited paper **Paper not presented

THE EWLMII, A MEASUREMENT AND DATA ACQUISITION SYSTEM FOR RADON-DAUGHTERS

D.J. Keefe, W.P. McDowell and P.G. Groer*

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INTRODUCTION

Historical Background[1,2,3]

In 1972, the Radiological and Environmental Research Division of Argonne National Laboratory contracted with the United States Bureau of Mines for the development of three identical instruments to measure Rn-daughter concentrations and Working Level (WL), in uranium mine atmospheres. This project, under the direction of Dr. Peter Groer, led to the development of the Instant Working Level Meter (IWLM). The basic principles for the Groer method of measurement of Rn-daughter concentration were developed and proven with these instruments. These IWLM's were small portable units uitlizing a calculator chip to automatically make the mathematical calculations required to compute the WL. CMOS circuitry and a small 12 liter/minute pump were used to permit battery powered operation.

This development let to another contract between the Electronics Division of Argonne National Laboratory and the Bureau of Mines for the development of the Remote Working Level Monitor (RWLM). The RWLM's electronics package was micro-processor based to provide automatic operation. The monitor was programmable via a keyboard to start and stop at selected times and to take measurements at desired time intervals, providing a fully automatic operation. This instrument differed from the IWLM in several ways:

It was a stationary unit with two remote monitoring stations automatically controlled from a central micro-processor. It was similar in its measurement sensitivity, (.01 WL) and utilized a flow rate of 12 liters per minute. The RWLM, however, contained an automatic filter advance mechanism where the IWLM was manually operated. The data were printed out on a Texas Instruments Silent 700 terminal.

The Environmental Working Level Monitors (EWLM) were developed under a contract between the EPA and ANL. Under this contract, four instruments were built. Although the method of measurement was the same for all instruments developed, these units differed considerably from the previously constructed instruments. The first difference between the EWLM and the RWLM was measurement sensitivity. The EWLM had a sensitivity of (.001 WL) and a sampling flowrate of approximately 40 liters per minute compared to (.01 WL) and 12 liters per minute for the RWLM. The second significant difference was that each EWLM was self contained. That is, it contained a complete system, including the micro-computer in one portable aluminum case. The EWLM's were designed for environmental measurements in non-mine atmospheres.

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*Institute for Energy Analysis, Oak Ridge, Tenn.

As experience with the field operation of the EWLM's accumulated, the ease with which these instruments automatically monitored environmental Rn-daughter concentrations and WL became more and more evident. This field experience led to the development of additional EWLM's for two of the scientific divisions at Argonne National Laboratory. The new EWLM's incorporated improvements designed to correct some maintenance problems which occurred in the original EWLM design. These new instruments (EWLMII) and their differences with the EWLM's are discussed in detail below.

GROER METHOD[1,2,3,4]

A brief review of the Groer method for the measurement of the Rn-daughter concentrations and the WL is beneficial in the understanding of the operation and principles of the EWLM and EWLMII.

The EWLM and the EWLMII use identical timing intervals to accumulate the data for the Groer method of Rn-daughter measurement. They first take a background count for three minutes. They then collect an air sample onto a filter membrane for three minutes and then move the filter membrane to a counting station. This air sample is then counted for three minutes starting 13 seconds after the end of the sampling period. The instrument has three counting channels; the lower-energy alpha channel, the upper-energy alpha channel, and a beta sensitive channel. The RaA counts observed are accumulated in the lower-energy alpha channel; the RaC' counts are stored in the upper-energy alpha channel, which the total beta counts from RaB and RaC are recorded in the beta channel. The counts in these three channels are functions of N_A, N_B, N_C, the unknown concentrations of RaA, RaB and RaC in the ambient air (units are atoms/liter). The following relationships hold:

$$\begin{aligned} TA &= 1.023403\ E_1 V N_A \\ B + C &= (0.110393E_2 + 0.007653E_3) V N_A + (0.214298E_2 \\ &\quad + 0.023559) V N_B + 0.283042 E_3 V N_C \\ TC' &= (0.007653N_A + 0.023559N_B + 0.283042N_C) E_1 V \qquad (1) \end{aligned}$$

where:

TA = alpha counts from RaA accumulated during the 3 min counting period starting 13 sec after the end of sampling.

B + C = beta counts from RaB and RaC in beta channel accumulated during the same time interval as above.

TC' = alpha counts from RaC' (same period of accumulation as above).

V = flowrate (liters/min).

E_1 = detection efficiency from RaA and RaC'.

E_2 = detection efficiency from RaB.

E_3 = detection efficiency from RaC.

N_A = RaA concentration in units of atoms/liter.

N_B = RaB concentration in units of atoms/liter.

N_C = RaC concentration in units of atoms/liter.

The numerical coefficients in (1) follow from the laws of radioactive series decay. The half-lives used are:

Nuclide	Half-life
RaA (^{218}Po)	3.05 min
RaB (^{214}Pb)	26.8 min
RaC (^{214}Bi)	19.7 min

To show the principle of calculation of these numerical coefficients, a sample calculation is given below:

EXAMPLE: Calculate the numerical coefficient for the first of (1).

This coefficient is the product of three factors:

1) Buildup factor = $1 - \exp(-\lambda_A t_B)$

t_B = buildup time = 3 min.

2) Delay factor = $\exp(-\lambda_A D)$

D = Delay before start of the counting interval

= 13 sec or 13/60 min.

3) Decay factor = $[1 - \exp(-\lambda_A t_D)]\lambda/A$

t_D = decay time = 3 min.

λ_A = RaA decay constant = 0.2272614.

Multiplying all these factors one obtains:

$$[1 - \exp(-\lambda_A t_B)]\exp(-\lambda_A D)[1-\exp(-\lambda_A t_D)]/\lambda_A = 1.023403.$$

The derivations of the analogous coefficients for the other equations are more complex but easily obtainable by the computer programs. If all the efficiencies and the flow rate are known, (1) contains only numerical coefficients and the unknowns N_A, N_B and N_C. Inverting (1) results in another set of equations which give a N_A, N_B, and N_C for every observed set of TA, (B+C) and TC'. The WL can be calculated easily from the resulting N_A, N_B, and N_C.

The alpha efficiency E_1 is easily measured in the laboratory, but to determine the beta efficiencies, E_2 and E_3, a calibration run must first be made. Once E_1 is known, E_2 and E_3 can be determined by the following procedure. Knowledge of E_1 allows a complete analysis of any air sample by a method using total alpha counts. This method allows calculation of N_A, N_B and N_C for this particular air sample. N_A, N_B, and N_C can then be used to calculate the beta disintegrations from RaB and RaC. By comparing the calculated beta disintegrations with the observed total beta counts, the values for E_2 and E_3 are obtained. The necessary calculations are all performed by the "EWLM CALIBRATION PROGRAM."

To calculate the beta efficiencies E_2 and E_3, the Rn-daughter concentrations (atoms liter) N_A, N_B and N_C must first be determined from the alpha counts during several counting periods using the following equations:

$$N_A = 0.711318*A_5/E_1*V$$

$$N_B = (-.689775*A_5 - 7.54536*C_5 + 1.86689*C_3)/E_1*V$$

$$N_C = (0.0441761*A_5 + 3.01566*C_5 - 0.203315*C_3)/E_1*V \qquad (2)$$

where:+

A_5 = RaA counts observed during 5 min starting 13 sec after the end of the 3 min sampling time.

C_5 = RaC' counts observed during the same time interval as above.

C_3 = RaC' counts observed during 30 min. starting at the same time as above.

The numerical coefficients in (2) are again derived from the laws of radioactive-series decay. This derivation is straight-forward but lengthy and will therefore be omitted.

With N_A, N_B and N_C known, E_2 and E_3 can be determined from the following equations:

+The overlap corrections must be incorporated into the alpha counts (A_5, C_5 and C_3).

$$Q_1 = (0.212378*N_A + 0.348201*N_B)*V*E_2 + (0.019829*N_A + 0.0496395*N_B + 0.455804*N_C)*V*E_3$$

$$Q_2 = (1.482571*N_A + 1.549219*N_B)*V*E_2 + (0.599567*N_A + 0.736276*N_B + 1.842208*N_C)*V*E_3 \qquad (3)$$

where:

Q_1 = total beta counts observed during 5 min starting 13 sec after the end of the 3 min sampling time.

Q_2 = total beta counts observed during 30 min same time as above.

Inverting equations (3), the beta efficiencies E_2 and E_3 can be determined, then (1) can be inverted and properly scaled. When properly scaled, this yields a set of equations which gives the Rn-daughter concentrations in pCi/liter. The WL can also be expressed as a linear combination of A, (B + C) and C'. These counts are net counts, i.e., the background has been subtracted. The equations are of the following form:

$$\begin{aligned} WL &= C_1(A) + C_2(B + C) + C_3(C') \\ RaA(pCi/liter) &= C_4(A) + C_5(B + C) + C_6(C') \\ RaB(pCi/liter) &= C_7(A) + C_8(B + C) + C_9(C') \\ RaC(pCi/liter) &= C_{10}(A) + C_{11}(B + C) + C_{12}(C') \end{aligned} \qquad (4)$$

C_1 through C_{12} are the derived weighting coefficients which are stored in the memory of the EWLM. It is clear from this description of the calibration, that N_A, N_B and N_C are treated as independent unknowns (i.e., no *a priori* relationship between these quantities other than radioactive series decay is assumed). The EWLM determines, therefore, the Rn-daughter concentrations and WL, without any assumptions about the Rn-daughter equilibrium.

The EWLMII

Calibration[5,6]

To be able to measure the WL and the Rn-daughter concentrations, the coefficients C_1 through C_{12} of equations (4) must be contained in the EWLMII's permanent memory. A calibration program automates the solution of the equations for radioactive-series decay, as applied to the EWLMII, and calculates the required efficiencies and coefficients. However, to provide this external program with input data, a calibration run by the EWLMII is required first.

An internal calibration program is requested via the keyboard which automatically takes a series of 20, 41-minute calibration runs. In each 41-minute run a normal measurement is made using a single 3-minute sample but the counting time is not terminated after 3 minutes as in a regular run. Data is stored in memory and printed out on the printer for several time periods. The data printed out is the WL followed by the Rn-daughter concentrations for a standard run as well as the counts for all the counting channels for 3 minutes, 5 minutes, 30 minutes, 35 minutes and the counts during the time interval between 39 and 41 minutes after sampling.

This data can then be used as input for a "Calibration Program" running on a separate microcomputer. After entering the data generated by the calibration runs of the EWLMII into this external program, it will perform the following functions:

1) It calculates the Rn-daughter concentrations by three different methods and the WL by four different methods from the input data of each individual run. The four different methods are, the alpha-spectroscopic method, the total-alpha method, the Kusnetz method and the Groer method.

2) It calculates the RaB and RaC efficiencies using the Rn-daughter concentrations calculated by the total-alpha method, and the beta counts at two different periods of time.

3) It calculates the 12 coefficients needed for the Groer method.

4) It calculates the Rn-daughter concentrations from 9 of these calculated coefficients, in units of atoms/liter.

5) It calculates the WL from the Rn-daughter concentrations, and with the use of the three remaining coefficients it also determines the WL directly from the measured counts.

This calibration program has two branches, one which calculates the beta efficiencies from the input data, and another which uses the mean value of the beta efficiencies previously calculated to derive the final weighted coefficients. A tabulation of the beta efficiency values calculated in 20 separate calibration runs and the final weighted coefficients are shown in the Table of Efficiencies and Coefficients.

Inputting the mean value of the beta efficiencies into the second branch of this program, as well as the input count data asked for by the program, results in the calculated values of the Rn-daughter concentrations and WL. The calculated concentrations and WL's are for a single 3 minute sample of air.

A summary of the calculated values is shown in the Table of Calibration Data. The units used in this table for the Rn-daughters are Atoms/Liter. The constants used in these calculations for the specific Unit calibrated are also shown, with, EA the alpha efficiency, EB and EC the RaB and RaC beta efficiencies respectively, V the flow-rate in liters/minute and OL the overlap correction factor. You will notice good agreement between the values of the Rn-daughter concentrations and the WL's calculated by the various independent methods for each individual run.

Electronic and Mechanical Improvements

After extended use of the EWLM's in the field, certain areas of high maintenance began to appear. To eliminate these problems modifications were incorporated into the EWLMII and are listed below:

The Bomar printer suffered from excessive mechanical failures because its plastic parts were apparently too fragile for continuous operation. Therefore, the printer and its interface card were replaced with a more reliable system. A Texas Instruments model EPN 9120 thermal printer and a standardized RS-232 serial interface card were substituted.

The take-up gear assembly on the automatic filter advance mechanism required proper lubrication and adjustment and was prone to failure if not maintained properly. This caused failures in the filter advance cycle. To solve this problem, a torque motor was substituted for the take-up gears to provide the desired filter advance operation.

The paper position fiber-optic sensor caused failures in the filter advance cycle because the optical sensitivity of the sensor was too great. This occasionally caused the filter advance to prematurely stop due to light reflections received 180 degrees out of phase with the positioning mechanism. The solution to this problem was to use a different optical sensing method which eliminated the fiber-optics and used a less expensive, less sensitive, directly coupled optical sensor.

The solenoid clutch-brake occasionally failed due to binding of its cylindrical shaft which prevented full closure of the brake assembly. To eliminate this problem a complete redesign of the clutch-brake assembly was made. This included a longer cylindrical shaft on the clutch assembly and additional windings in the coil assembly.

The scotch yoke assembly of the filter transport mechanism showed excessive wear due to the lack of proper lubrication. The solution was to install a sealed bearing on the driver shaft.

To adjust the length of movement for precise filter membrane positioning in the EWLM's, shimming of the top plate of the filter transport mechanism was required. A more flexible positioning method was needed. Therefore, we redesigned the upper roller arm of the paper drive assembly incorporating a locked cam adjustment in order to more conveniently adjust the filter membrane positioning.

In the process of servicing the EWLM's, we found that they frequently needed to be removed from the outer case. This process was found to be time consuming and costly as all sub-assemblies were bolted separately to the outer case. Therefore, we decided to repackage the EWLMII to provide easy access to all parts without requiring disassembly. An inner mounting frame was designed to which all components could be fastened and which could easily be removed from the outer case. This allowed easy access to components for troubleshooting and repair.

On extended monitoring periods we found that the printer paper was occasionally blocking the airflow into the sample port. Therefore, the filter advance mechanism was repositioned with respect to the printer so as to eliminate any possibility of obstruction of the air sampling port.

In addition to the modifications for the EWLMII listed above, certain other improvements were incorporated. The standard (STD) bus was selected to implement the electronics package. This allowed us to purchase commercially most of the required micro-processor cards such as the CPU, the serial and parallel I/O cards and the memory card. We updated our CPU from the 8080 to the 8085 which reduced power requirements and doubled the processing speed. The PROM memory capacity was doubled by using 2k memory chips rather than the original 1k memory chips.

A 16 channel analog to digital converter card has been incorporated in order to be able to measure additional parameters. This card, along with the addition of a digital data recorder allows us to transfer data more efficiently from the EWLMII to external computers for data analysis.

Software Changes

The original operating programs for the RWLM and EWLM were written in PL/M, a high level language developed by the Intel corporation. At that time, Intel's software consisted of a compiler and cross assembler residing externally to their development system. This meant that the compiling and loading of the hex files had to be made via a phone linked into TSO, a time sharing program used by our central computer facility. Although the compiler allowed structured programming it had no capability to link assembly language programs and provided non-relocatable addressing. With these restrictions we were forced to resort to tricks to use assembly language modules such as a floating point mathematics package. To make a small change in the program, the entire program had to be recompiled. This was time consuming both in the compiling operation as well as the resulting debugging that was often required.

However, with the introduction of a resident software PL/M the Intel package now provides linking and locating capabilities. This allowed us to modularize the programming of the EWLMII, linking and locating modules in our local development systems. By building libraries of these modules linking was further simplified. Now minor changes to the program can be made at the module level, and only those modules changed required recompiling. This also means that new modules could quickly be written and linked into the main program. With the use of this new software package, the new or modified modules could then be linked and located in minutes rather than the days taken in the past to recompile and debug the program.

Updated PROM programmers further reduced the time required to change memory. These new instruments and software allow us to down line load several modules at a time into the EWLMII's RAM memory for debugging. After all modules have been thoroughly checked out, the entire program is re-linked and loaded into the PROM programmers buffer. Each PROM for the EWLMII's operating system is then programmed and inserted into the memory board. This process greatly accelerates the time needed to debug, check out and program a new operating program.

MEASUREMENT OF ADDITIONAL PARAMETERS

The increased memory capacity and the addition of an Analog I/O subsystem in the EWLMII allows us to measure additional parameters. An Analog Devices Model RT-1225 STD bus compatible card was used. It provides the system with 16 channels of analog input and 2 channels of analog output. It contains a standard sample and hold amplifier with a monolithic 10 bit A/D converter with the flexibility of 16 single ended or 8 differential inputs. It operates off a single +5 volt power supply and contains a memory mapped I/O.

Through the use of this card additional parameters such as, inside and outside temperatures, atmospheric pressure, wind velocity and direction, humidity and control switches are interfaced to the EWLMII, making this instrument a true data acquisition system. These external DC inputs and others can be added to the system to output additional parameters on either a predetermined or programmable schedule. Additional program modules for this added capability can easily be linked into the main EWLMII program. This relinked program can then be burned into the PROM's to provide this additional monitoring ability.

Data Storage, Display and Analysis

The EWLM and EWLMII temporarily store their data in active memory (RAM) and this information is then transferred to the printer at the completion of the measurement cycle. Subsequent measurements use the same data area causing the loss of previous data. The only storage media used in the EWLM was the printed data stored on a continuous paper tape. This method of data readout required that these recorded values be entered into an external computer for analysis.

To streamline the data processing, a TEAC MT-2 digital data recorder was added to the EWLMII. Immediately after the data has been printed out, it is then recorded on the digital cassette recorder. The EWLM's print out a short report onto the 20 character per line printer. There is additonal data stored in memory which is not printed out in this short report. The digital cassette records the complete memory image, therefore all data is permanently stored on the cassette. This allows us to use any part of or all of this data for our analysis. The data from each sample is continuously stored on the tape. When the series of runs is complete an "E" is entered into the keyboard which is then removed from the EWLMII and is ready for a data transfer.

To display and analyze this data stored on the cassette, we used a microcomputer with 32k of memory configured with an identical digital data recorder, an eight inch floppy disk and a dual serial port.

Software was written which loads the tape, transfers the cassette tape's data into the micro-computers memory and then rewinds the tape. After the data have been transferred to the computer's memory the cassette can be removed for reuse in the EWLMII. Another program was written to display a complete report of the data on the terminal or line printer. Additional programs are to be written for data analysis. This stored data can be transferred to the floppy disk and data from additional tapes can be added to this extended data file. This library of data can then be used for data analysis. Programs can be written to either tabulate or plot the WL and Rn-daughter concentrations versus time. Other manipulations of the data can be printed such as Rn-daughter ratios, statistical evaluations and comparisons of the daughter concentrations with: barometric pressure, humidity, temperature, inside-outside temperature differences, air circulation rates, air filter effects with air flow-rates and time and other parameters.

CONCLUSION

Development of new methods and instruments for the measurement of WL and Rn-daughter concentrations has been in progress for several years. However, developing efficient methods of data acquisition has not been the primary thrust of this research. If large volumes of data from automated instruments are to be efficiently analyzed, a built in data acquisition system is needed. One such instrument, the EWLMII, has been described in this paper. The need for accelerated research for the understanding and control of Rn-daughters can be enhanced by such an instrument.

References

1. P.G. Groer, D.J. Keefe, W.P. McDowell and R.F. Selman. "The Instant Working Level Meter." U.S. Bureau of Mines, Department of Interior, Final Report (August, 1974).

2. D.J. Keefe, W.P. McDowell and P.G. Groer. "The Remote Working Level Monitor." U.S. Bureau of Mines, Department of Interior, Final Report (November 18, 1977).

3. D.J. Keefe, W.P. McDowell and P.G. Groer. "The Environmental Working Level Monitor." United States Environmental Protection Agency, Final Report (September 29, 1978).

4. P.G. Groer, D.J. Keefe, W.P. McDowell and R.F. Selman. "An Instant Working Level Meter with Automatic Individual Radon Daughter Readout for Uranium Mines." Presented at the Third International Congress of the International Radiation Protection Association, Washington, D.C. (September 9-14 1973). Published in Proceedings (Part II: 950-956, National Technical Information Service).

5. W.P. McDowell, D.J. Keefe, P.G. Groer and R.T. Witek. "A Microprocessor-Assisted Calibration for a Remote Working Level Monitor". IEEE Trans. Nucl. Sci. NS-24, No. 1, 639-644 (February, 1977).

6. D.J. Keefe, W.P. McDowell, P.G. Groer and R.T. Witek. "Optimizing Techniques for the Measurement of Environmental Levels of Rn-Daughter Concentrations." IEEE Trans. Nucl. Sci. NS-25, 787-789 (February, 1978) Presented at the 1977 IEEE Nuclear Science Symposium, San Francisco, CA (October 19-21, 1977).

TABLE OF EFFICIENCIES AND COEFFICIENTS

EWLMII UNIT 2
2-9-81, TWILIGHT MINE URAVAN, COLORADO

RUN	EB	EC
1	.2020	.2887
2	.1799	.3129
3	.1769	.3167
4	.2146	.2772
5	.1954	.2983
6	.1574	.3374
7	.1835	.3017
8	.1949	.2850
9	.1565	.3345
10	.1784	.3053
11	.1663	.3177
12	.2076	.2746
13	.1915	.2874
14	.2247	.2455
15	.1684	.3116
16	.1565	.3230
17	.2203	.2505
18	.1676	.3078
19	.1703	.3147
20	.1914	.2771

AVERAGE EB = .1852 STANDARD DEVIATION = .0211

AVERAGE EC = .2984 STANDARD DEVIATION = .0250

COEFFICIENTS

4.7095478359E-06	3.49815313024E-05	-1.71403658529E-05	WL
6.36590042261E-03	0.00	-1.65513410988E-03	RaA
-3.73206192398E-04	7.52498502545E-03	-6.92698319685E-03	RaB
1.5610219182E-05	-8.52066462292E-04	5.28141146336E-03	RaC

TABLE OF CALIBRATION DATA
EWLMII UNIT 2
2-9-81, TWILIGHT MINE URAVAN, COLORDO

EA=.4028 EB=.1852 EC=.2984 V=39.01 OL=.26

		ALPHA SPEC.	TOT.ALPHA	GROER	KUZ
RUN 1					
	RaA	531.62	541.23	525.06	
	RaB	2234.72	2490.50	2511.20	
	RaC	1339.34	1281.56	1320.23	
	WL	.2671	.2798	.2816	.2759
RUN 2					
	RaA	502.94	509.71	502.62	
	RaB	2051.15	2237.05	2290.43	
	RaC	1280.54	1239.21	1259.60	
	WL	.2498	.2590	.2626	.2554
RUN 3					
	RaA	483.36	486.49	478.96	
	RaB	2032.74	2159.80	2143.99	
	RaC	1278.29	1254.74	1278.34	
	WL	.2465	.2529	.2526	.2544
RUN 4					
	RaA	476.93	489.85	474.01	
	RaB	1911.69	2077.52	2125.45	
	RaC	1225.43	1167.08	1226.23	
	WL	.2355	.2432	.2479	.2435
RUN 5					
	RaA	477.68	491.56	471.09	
	RaB	1877.39	2046.68	2084.18	
	RaC	1259.92	1198.17	1245.89	
	WL	.2356	.2434	.2463	.2527
RUN 6					
	RaA	476.76	466.65	461.54	
	RaB	1824.90	1885.25	1949.83	
	RaC	1176.87	1173.69	1156.76	
	WL	.2266	.2298	.2321	.2353
RUN 4					
	RaA	464.27	495.65	462.13	
	RaB	1853.58	2116.37	1951.37	
	RaC	1190.79	1064.14	1194.29	
	WL	.2287	.2401	.2345	.2355

EWLMII UNIT 2

		ALPHA SPEC.	TOT.ALPHA	GROER	KUZ
RUN 8	RaA	496.75	511.94	496.25	
	RaB	2071.82	2224.76	2062.23	
	RaC	1219.44	1155.39	1238.82	
	WL	.2467	.2536	.2472	.2563
RUN 9	RaA	492.43	483.54	487.69	
	RaB	1947.77	1984.61	1980.26	
	RaC	1244.22	1268.14	1264.01	
	WL	.2404	.2430	.2430	.2459
RUN 10	RaA	507.79	516.76	504.65	
	RaB	2092.49	2255.62	2126.36	
	RaC	1264.74	1219.06	1275.75	
	WL	.2518	.2597	.2541	.2582
RUN 11	RaA	514.50	523.17	517.78	
	RaB	2092.66	2186.74	2058.15	
	RaC	1249.48	1212.22	1263.51	
	WL	.2516	.2559	.2507	.2525
RUN 12	RaA	448.05	475.11	444.87	
	RaB	1814.10	2016.33	1836.98	
	RaC	1207.58	1101.00	1204.95	
	WL	.2256	.2342	.2265	.2231
RUN 13	RaA	493.76	582.21	492.39	
	RaB	1727.60	1955.32	1647.76	
	RaC	1175.96	1043.48	1214.78	
	WL	.2235	.2327	.2209	.2249
RUN 14	RaA	514.95	584.07	512.50	
	RaB	1700.60	2115.32	1774.62	
	RaC	1216.51	955.24	1218.41	
	WL	.2265	.2429	.2308	.2366
RUN 15	RaA	496.12	511.36	495.36	
	RaB	1813.69	1904.92	1703.11	
	RaC	1186.01	1128.48	1210.38	
	WL	.2294	.2330	.2242	.2317

EWLMII UNIT 2

	ALPHA SPEC.	TOT.ALPHA	GROER	KUZ
RUN 16				
RaA	447.41	455.23	445.48	
RaB	1694.11	1756.25	1479.86	
RaC	1059.00	1027.87	1105.09	
WL	.2097	.2124	.1996	.2126
RUN 17				
RaA	414.10	453.60	413.15	
RaB	1525.72	1751.83	1499.24	
RaC	1013.70	865.62	1020.21	
WL	.1936	.2023	.1923	.1917
RUN 18				
RaA	358.36	382.94	355.56	
RaB	1298.76	1396.09	1169.06	
RaC	817.77	730.37	840.17	
WL	.1627	.1659	.1561	.1607
RUN 19				
RaA	354.76	372.17	353.07	
RaB	1069.26	1143.29	957.81	
RaC	776.95	714.51	802.29	
WL	.1464	.1489	.1411	.1462
RUN 20				
RaA	358.06	378.46	358.52	
RaB	1145.07	1239.90	1044.75	
RaC	744.27	670.21	764.88	
WL	.1493	.1527	.1446	.1560

THE MEASUREMENT OF RADON-222 IN AIR BY DIRECT EXTRACTION INTO A LIQUID SCINTILLATOR

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ABSTRACT

Radon is extracted from ca. 10 liter ambient air samples directly into a solution of hexane and toluene held at -78 degrees C in a dry ice-acetone bath. The solution is sealed in a 22 ml glass liquid scintillation vial containing 2 ml of concentrated fluor. The vials are counted on a liquid scintillation counting system with a narrow counting window set around the alpha peaks of radon and its short-lived progeny. A large number of samples can be prepared in a relatively short time with minimal equipment, and counting is performed automatically. The large sample volume more than compensates for the relatively high alpha background of liquid scintillators. Alternative liquid scintillation sampling strategies are discussed.

INTRODUCTION

Limitations imposed by counting statistics require that accurate measurements of radon-222 in air at ambient concentrations (less than 1pCi/l) involve air samples of at least one liter. To meet this requirement, one must use either a large volume counting device or employ a concentration technique of some sort. The technique described here makes use of the high solubility of radon in certain organic solvents to extract radon from an air stream. If the solvent chosen is suitable for liquid scintillation counting, then the radon may be counted directly upon addition of appropriate fluors to the solution. The lower the temperature of the solvent at the time of extraction, the greater will be the degree of radon concentration and hence the sensitivity of the process. The temperature recommended for most applications is -78 C, the temperature of a dry ice and acetone bath. Several appropriate solvents are liquid at this temperature and dry ice is easily carried to remote locations in an ordinary ice chest. While this extraction process does not produce the complete radon retention obtained in techniques involving solid sorbents held at liquid nitrogen temperatures (e.g. Da73), the degree of extraction is reproducible and in good agreement with theory.

THEORY

The distribution of radon between air and a solvent in a closed system is described by L, the Ostwald coefficient, according to the equation:

$$L = \frac{C_s V_a}{C_a V_s} \qquad \text{(Eq. 1)}$$

where C_s and C_a are the equilibrium concentrations of gas in the solvent and air phases and V_s and V_a are the volumes of the two phases. The temperature dependence of the Ostwald coefficient for radon in various organic solvents is of the general form:

$$L = A + B \exp(-\upsilon\theta) \qquad \text{(Eq. 2) (We78)}$$

where θ is the temperature as expressed in corresponding degrees centigrade and A, B, and υ are experimentally determined parameters. Degrees of the "corresponding centigrade" scale are defined as one hundredth part of the interval between the melting and boiling points of the solvent in questions. The values of A, B, and υ for a number of solvents are listed in Table 1, and a graphical representation of L vs temperature is shown in Figure 1. Carbon disulfide, which has the highest affinity for radon in the temperature range shown, is a strong quenching agent and is incompatible with the liquid scintillation process. Normal hexane, however, is a workable liquid scintillation solvent (Ha53) and appears to be a better extractant than toluene, the more commonly used scintillation solvent. While the light output of a hexane-based scintillation solution is only a quarter that of a toluene-based solution, the high photon production associated with the alpha and hard beta emissions of radon and its daughters assures that ample light is available for detection.

If a sample of air with volume V_a and a radon concentration C is brought into equilibrium with a volume V_s of solvent having an Ostwald coefficient of L at the equilibrium temperature, then the activity residing in the solvent phase will be given by:

$$A_s = LV_sC/(1 + LV_s/V_a) \qquad \text{(Eq. 3)}$$

When the volume of air V_a is large compared to LV_s, A_s is relatively insensitive to variations in V_a, tending to a steady value of CLV_s as V_a tends to infinity. In this range, details of the contact process such as air flow rate and bubble size become unimportant. Figure 2 shows predicted radon retentions for three values of L and data from a calibration run which involved 20 ml of a solution consisting of 90% n-hexane and 10% toluene, by volume. At high air volumes the points fit the L = 300 curve, but fall below it as V_a becomes small compared to LV_s. For sample sizes of LV_s or greater, no flow rate dependence was observed between 0.5 and 3.0 liters per minute, and only a small reduction in radon retention was noted when an orifice that produced large bubbles was substituted for the regular frit that produced small bubbles.

At -78 C, the solubility of oxygen in organic solvents is considerably higher than at room temperature. Because oxygen reduces the light output of a scintillation solution, some consideration must be given to the effects of the additional oxygen acquired during radon extraction. While reduced light output itself is easily dealt with by a gain adjustment, uneven oxygen uptake from sample to sample could produce variations in counting efficiency. If the counting window is set to include the radon and polonium alpha peaks over a reasonable range of quench factors this problem can be minimized, as beta continuum counts lost on one side of the alpha peaks are approximately compensated for by the counts gained on the other as the peak centroids are shifted by varying degrees of quench. The external standard feature of most commercial liquid scintillation units can readily be used to determine whether a given sample lies within the acceptable quench range. Figure 3 shows two radon spectra near the extremes of quench that we have noted in practice. In this range, the variation in counting efficiency, as measured by the ratio of counts in the window to the total counts, is acceptably small. The quench due to absorbed oxygen can in fact be exploited to detect improperly sealed vials. The loss of oxygen due to a slow leak would decrease the quenching effect and raise the external standard ratio. A quick recount of the samples for external standard ratio performed 48 hours or more after the original count would be sufficient to indicate any vial that had lost oxygen, and in all probability, radon. In many cases, a recount of the "leakers" could then establish an effective half-life of radon in the vial, permitting the estimation of radon content at the time the vial was sealed.

EXPERIMENTAL METHOD

The choice of extraction temperature and primary solvent represents a trade-off between convenience and completeness of radon recovery. As figure 1 demonstrates, the lower the temperature, the more efficient the extraction will be. The higher the efficiency, the larger the air sample that it is practical to process, as Figure 2 shows. In effect then, it is the size of the sample required for the desired degree of sensitivity that controls the maximum practical extraction temperature. For the measurement of radon at ambient outdoor concentrations, the -78 C extraction into hexane as shown in Figure 2 represents an acceptable balance between sensitivity and convenience. The dry ice required for the low temperature bath is inexpensive and easily carried in ordinary styrofoam or plastic containers. The extraction train shown in Figure 4 (not to scale) is composed of standard laboratory equipment. Collapsible 20 liter sample bags (A) are filled at locations of interest and returned to a central field station for processing. If the time between sampling and processing is more than an hour or so, as would be the case in a 24 hour integrated bag sample, care must be taken that the bag is composed of material that has been shown to be impervious to radon. At the extraction station, air is drawn from the bags by a pump (B) at approximately 2 liters per minute through a dessicator or cold trap (C) to remove moisture that might condense and block portions of the sample train. The dried air passes through an impinger tube (E) containing 20 ml of solvent held at -78 C in the dry ice and acetone bath (D). The airstream is broken into fine bubbles by the fritted orifice of the impinger, thus assuring good phase contact between the air and the solvent. After warming to near ambient temperature, the exhaust air passes through a flow meter (F). When the desired volume of air has been passed through the solvent, the pump is stopped, the impinger disconnected, and its contents gently poured into a pre-chilled vial containing 2 ml of concentrated fluors (40 g/l PPO and 5 g/l dimethyl POPOP, or similar commercial concentrate.) The vial is then tightly capped with a plastic cone-style cap and placed in an ice bath to warm to 0 C. The impinger tube is then rinsed and 20 ml of cooled solvent is added in preparation for the next extraction. (Note - the thermal expansion of hexane over a 100 C interval is not trivial.) After a batch of vials has warmed to 0 C in the ice bath, each cap is briefly (ca. 1 second) loosened to relieve excess pressure, then tightened again. This step is a precaution against vial breakage due to overpressure and causes less than 1% radon loss. The vials are then allowed to warm to room temperature, and are ready for counting at any time later than three hours after initial sealing.

PRECISION AND SENSITIVITY

Each picocurie of radon-222 in equilibrium with its short-lived progeny produces on the average 11.1 alpha or hard beta disintegrations per minute. A window set as shown in Figure 3 wil accept approximately 80% of these events, or 8.9 cpm. Equation 2 predicts that 37.5% of the radon in 10 liter air sample will be retained in a 20 ml solvent phase with L=300, so that 33.4 counts per pCi/l of radon-222 are expected in the counting window at full equilibrium.

Because alpha particles are relatively inefficient light producers in liquid scintillators, energy discrimination between alphas and betas is not possible and compton electrons from environmental gamma radiation contribute heavily to the background in the region of the radon and polonium alpha peaks. In the window shown in Figure 3 the background will be approximately 12 cpm, depending on the exact window settings and the performance of the counting system. If, for example, the concentration of radon in air is 0.25 pCi/l, a 100 minute count on a 10 l sample will produce 825 expected net counts against an expected background of 1200 counts. The minimum detectable true activity (Al63) would be 0.04 pCi/l if the lack of independence of radon daughter counts can be neglected. Although the background is considerably higher than would be obtained from a Zn:S phosphor, it is seen that the large effective sample volume provides sufficient compensaton for radon concentrations encountered in continental atmospheres and in dwellings. In a recent intercomparison at the Department of Energy's Environmental Measurements Laboratory (Ge81), four sample bags were filled with air from the radon chamber and shipped to our Houston Laboratory for processing. The mean of the four samples was 46.2 pCi/l with a standard deviation of 0.8 pCi/l, as compared with the nominal value of 44.6 = 1.1 pCi/l.

ALTERNATE STRATEGIES

A great deal of latitude exists in the choice of solvent, the operating temperature, sample volume, and solvent volume. (Twenty milliliters is, however, an effective upper limit to the solvent volume if a commercial liquid scintillation counting system is to be used.) Some common solvents have liquid ranges that permit their use at temperatures well below -78 C. Pentane is liquid in the range -130 to 36 C, and isopentane is liquid in the interval -160 to 28 C. If nearly complete radon recovery from large air samples is desired, a low freezing point solvent cooled by an appropriate bath or device could be used in place of the -78 C system described above. If the sample volume were less than LV_S, however, it is probable that the details of phase contact would influence the degree of sample retention much more seriously than in the -78 C case.

In environments with radon concentrations appreciably above normal background it would be possible to operate effectively at 0 C or at ambient temperature. The quantity LV_S would then be relatively small, on the order of 500 ml. At air sample volumes appreciably above that, the concentration of radon in solvent would tend to LC. Weigel (We78) states that olive oil and other hydrocarbons of vegetable origin have Ostwald coefficients of about 28 at room temperature, which implies that 20 ml of solvent in equilibrium with air would contain about the same amount of radon as a conventional 500 ml grab sample. Whether or not ambient temperature extraction is a desirable alternative to conventional grab sampling would depend upon the logistics of the individual sampling problem.

The high affinity of organic solvents for radon leads to a simple and sensitive method for detecting radon that has been entrained on an activated carbon trap. Activated carbon at the temperature of dry ice is easily used as a radon trap, but the subsequent removal of the radon is somewhat more complicated, and requires elevated temperatures, vacuum, and pressurized gas. We have noted that 98% of the radon can be removed from the carbon simply by sealing the carbon in several volumes of a solvent such as toluene or hexane for a few hours, after which the radon can be counted by liquid scintillation. To confirm the degree of desorption, one gram of activated carbon upon which radon had been entrained at -78 C was divided into equal portions after thorough mixing. One portion was immediately suspended in 20 ml of scintillating gel and the other portion was placed in 15 ml of toluene for 3 hours. The toluene was then decanted and the carbon rinsed with an additional 5 ml of toluene, after which the carbon was suspended in a gel in the same manner as the first portion. While alpha particles emitted by radon and progeny within the carbon granules could not be expected to contribute effectively to the count rate, the energetic betas emitted by Pb-214 and Bi-214 did register well in the gel suspension. It was found that the carbon fraction that had been treated with toluene displayed less than 2% of the count rate of the fraction that had been placed directly in the gel.

The potential problem of radon loss during the initial second or two of desorption was solved by placing the cold carbon granules directly into a vial containing 10 ml of cyclohexane that had been frozen by placing the vial in the dry ice container. The vial was then tightly capped and allowed to come to ambient temperature. At some convenient time after the carbon had sunk to the bottom of the liquid cyclohexane, the vials were placed back in the dry ice container until the cyclohexane was again frozen. Ten ml of toluene scintillation fluid was then added to the vial and the vials were recapped and allowed to warm to room temperature. At this stage, the solvent could be withdrawn for counting, or counting could proceed with the carbon granules resting on the bottom of the vial. When 1.5 g carbon traps were used, the counts lost due to the presence of the carbon were found to be a small and reproducible fraction of the total.

CONCLUSION

The high solubility of radon in various liquid scintillation compatible solvents leads to a set of simple and sensitive concentration techniques. The choice of solvent, operating temperature, and sample volume can be varied in response to a wide range of sampling requirements. The relatively high background associated with liquid scintillators is overcome by the high sample volume in most applications. The liquid scintillator extraction techniques seem most promising for applications in which a large number of samples are routinely processed, such as a survey of indoor radon levels in a community. Table 2 shows data obtained by this technique from dwellings in Maine compared with data from commercial alpha track detectors (c.f. Pr81). The track detectors provide a measure of the integrated radon "exposure" over periods of three to six months, while the grab samples indicate only the radon concentration at the time of the alpha track detectors' deployment. Nevertheless, the descriptions of the radon levels in the community obtained by the two methods are quite similar. The means of the indoor measurements are not statistically different from the mean of the grab samples, as shown by the p-values of a two sample t-test, and the grab and alpha track measurements are correlated at the $p = .01$ level. The outdoor and basement alpha track data do not show the same agreement with the indoor grab samples, of course. We do not suggest that grab samples by this or any other method are an acceptable substitute for integrated measurements in all cases. If due consideration is given to meteorological variables, however, reasonable estimates of a community radon profile can be made with grab sampling techniques which offer the investigator the advantage of low cost along with prompt and controlled access to the data.

References

Al63 Altshuler, B. and Pasternack, B, 1963, "Statistical Measures of the Lower Limit of Detection of a Radioactivity Counter", Health Physics 9, 293.

Da73 Darrall, K.G., Richardson P.J., and Tyler, J.F.C., 1973, "An Emanation Method for Determining Radium Using Liquid Scintillation Counting", Analyst 98, pp. 610-615.

Ge81 George, Andreas, and Fisenne, Isabel, personal communication, 1981.

Ha53 Hayes, F.N., Rogers, B.S., and Sanders, P.C., 1953, "Importance of Solvent in Liquid Scintillators" Nucleonics 13, pp. 46-48.

Lo28 Loomis, A.G., 1928, "Solubilities of Gases in Liquids" in International Critical Tables of Numerical Data Vol. III, pp. 254-271, McGraw-Hill, New York.

Pr81 Prichard, H.M., Gesell, T.F., Hess, C.T., Weiffenbach, C., and Nyberg, P., 1981, "Integrating Radon Detector Data from Dwellings in Maine and Texas", submitted to Health Physics.

We78 Weigel, F., 1978, "Radon", Chemiker Zeitung 102, pp. 287-299.

Table 1

Solubility Coefficients for Radon in Various Solvents*

Solvent	Melting Point (°C)	Boiling Point (°C)	A	B	V
CS_2	-110	46.3	9	880	.051
n-Hexane	-95	68	5	345	.050
Ethyl Ether	-116.2	34.6	10	700	.055
Toluene	-92.4	110.7	2	125	.045
Acetone	-94.6	56.1	4	68	.046
Ethanol	-117.3	78.5	2.5	86	.046

*Values taken from Weigel (We78) except for those of n-Hexane, which were obtained by regression from the data of Loomis (Lo28).

Table 2

Agreement Between Indoor Grab Samples and Integrated Alpha Track Measurements in Maine

Sample Type	n	GM (pCi/l)	GSD	P-value of t-Test	Correlation Coefficient
First Floor Grab	57	1.67	3.0		
Alpha Track					
Bathroom	81	1.62	2.4	0.88	0.74 *
First Floor	82	1.40	2.4	0.33	0.71 *
Bedroom	81	1.12	2.4	0.17	0.73 *
Basement	77	2.46	2.4	0.04	0.46 *
Outdoor	67	0.46	2.4	0.00	0.15 *

*p less than 0.01

FIGURE CAPTIONS

Figure 1: Extrapolated Radon Solubilites (L) in Various Solvents as a Function of Temperature.

Figure 2: Predicted Radon Retention in 20 ml of Solvent in Equilibrium with a Range of Air Volumes as a Function of Solubility (L). Triangles Indicate Experimental Values.

Figure 3: Spectra of Radon-222 in a Hexane-Toluene Solution at Two Quench Levels. At the Indicated Window Setting, the Count Rate is Nearly Independent of Quench Within the Range Shown.

Figure 4: Diagram of Extraction Train (Not to Scale).

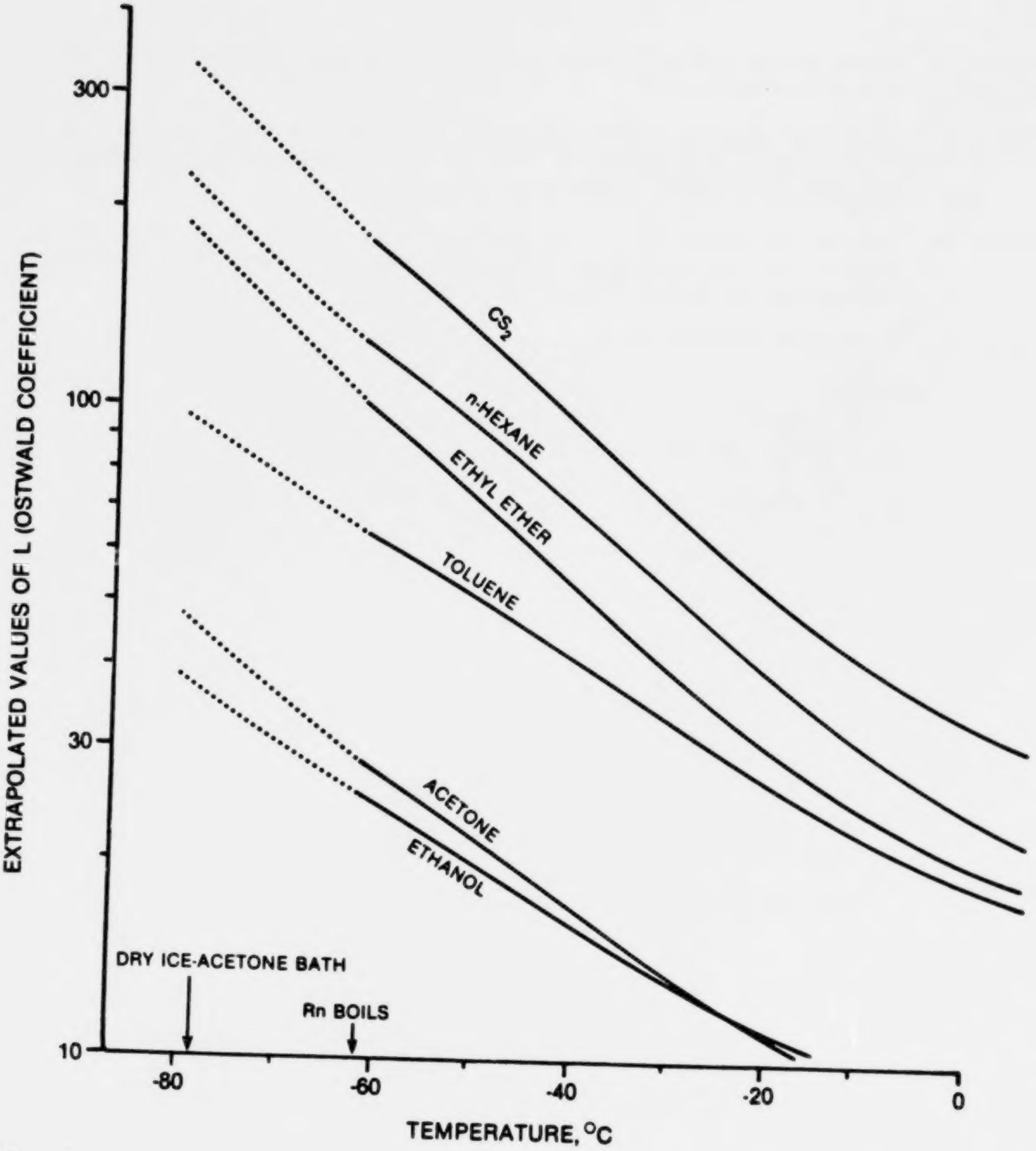
EXTRAPOLATED VALUES OF L (OSTWALD COEFFICIENT)
300
100
30
10
CS$_2$
n-HEXANE
ETHYL ETHER
TOLUENE
ACETONE
ETHANOL
DRY ICE-ACETONE BATH
Rn BOILS
-80
-60
-40
-20
0
TEMPERATURE, °C

Figure 1

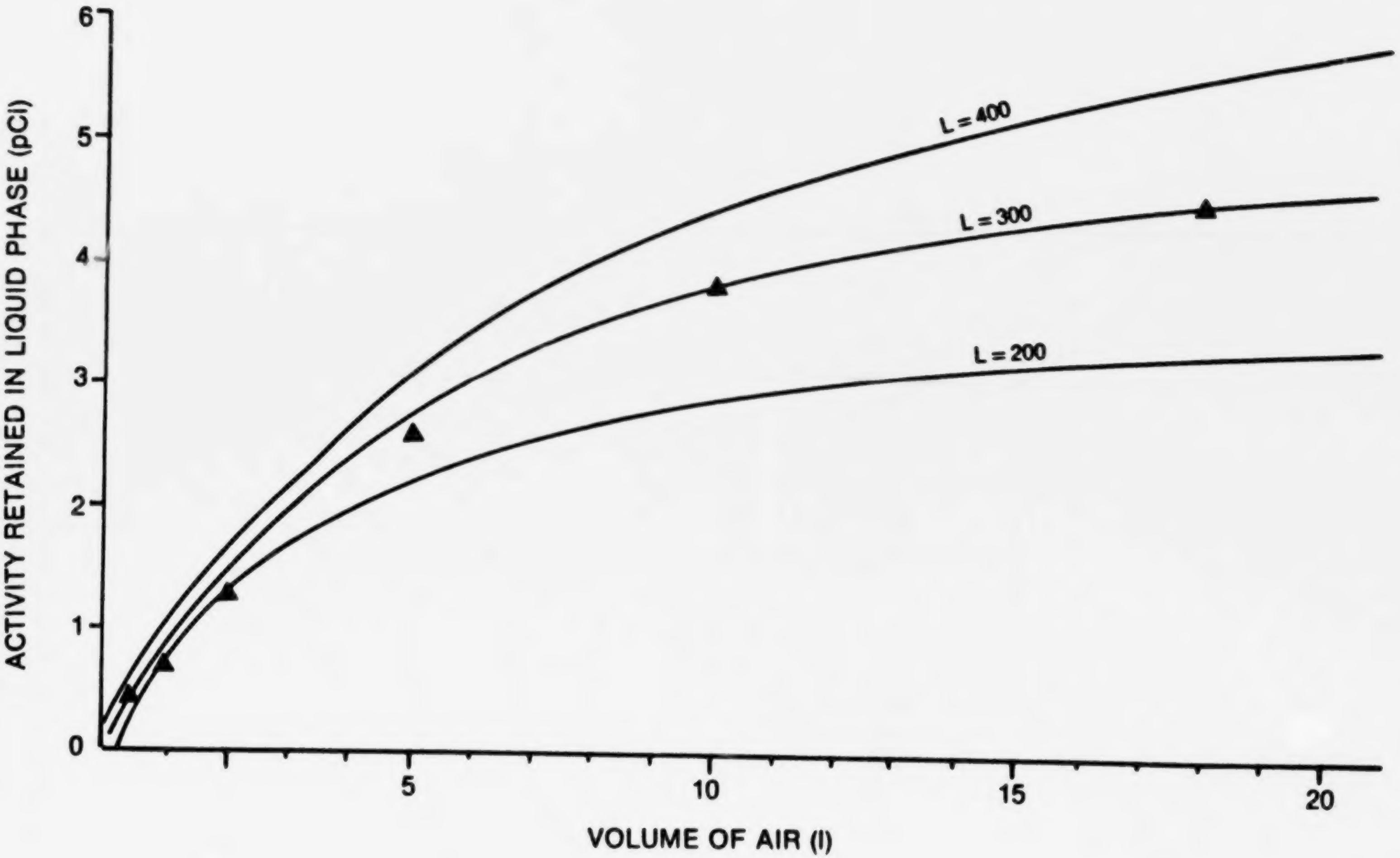
ACTIVITY RETAINED IN LIQUID PHASE (pCi)
0
1
2
3
4
5
6
VOLUME OF AIR (l)
5
10
15
20
L = 400
L = 300
L = 200

Figure 2

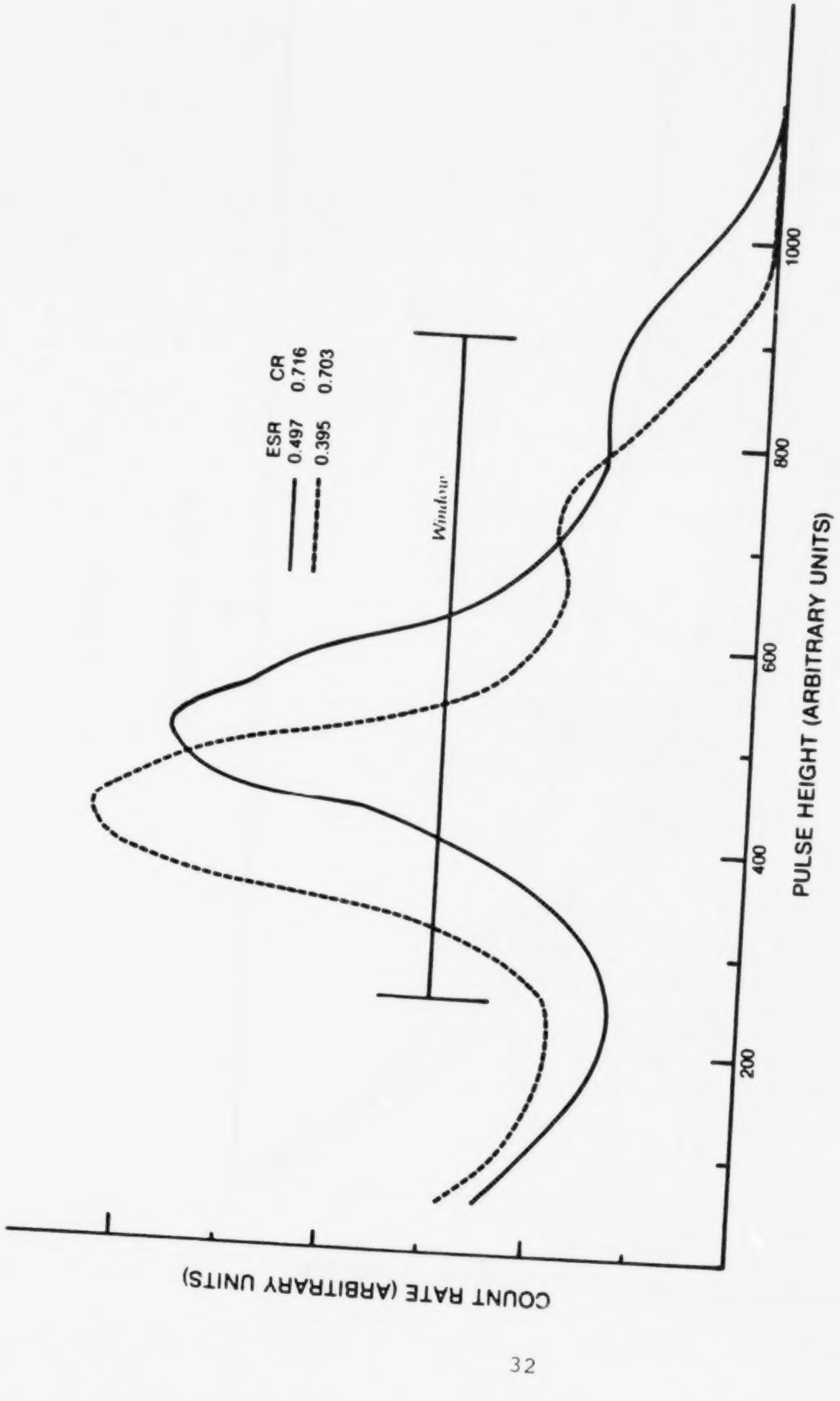
COUNT RATE (ARBITRARY UNITS)
PULSE HEIGHT (ARBITRARY UNITS)
200
400
600
800
1000
ESR CR
0.497 0.716
0.395 0.703
Window

Figure 3

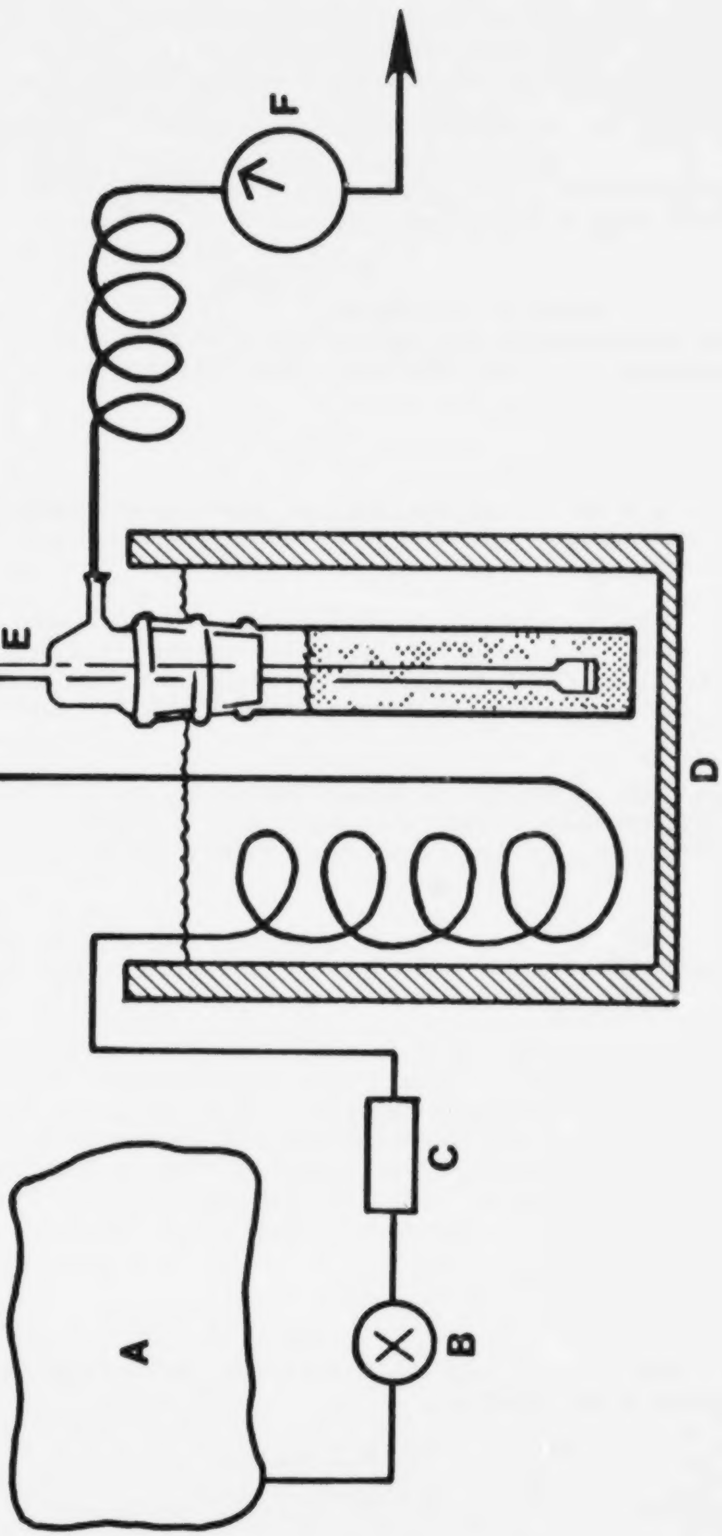

Figure 4

APPLICATION OF SURFACE-BARRIER DETECTORS FOR THE MEASUREMENT OF ENVIRONMENTAL RADON AND RADON DAUGHTERS IN AIR

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ABSTRACT

Surface-barrier detectors have been applied for alpha-spectroscopic measurements of environmental levels of airborne radon (Rn-222) and thoron (Rn-220) as well as for their individual short-lived daughters.

For the purpose of radon and thoron detection, the air has been dried, filtered and sucked into an evacuated aluminum sphere (14.2ℓ). The positively charged Po-218 (RaA)- and Po-216 (ThA)- atoms are collected on a surface-barrier detector by an applied positive voltage of 20kV between sphere and detector surface.

The alpha disintegrations of Po-218 (6.00MeV) and of Po-216 (6.77MeV) are counted separately by a field multichannel analyzer. Taking a one hour counting time at least 0.03 pCi/ℓ radon can be measured within 20% of statistical accuracy.

The same sensitivity is achieved using a flow-through version of that instrument for continuous operation. For thoron the sensitivity is only 1 pCi/ℓ corresponding to a flow rate of 1.6ℓ/min.

Using a separate instrument radon and thoron daughters are collected on a membrane filter and the decays of Po-218 (RaA), Po-214 (RaC') and Po-212 (ThC') are counted by means of alpha-spectroscopy during and after sampling. Good results have been obtained for a counting/sampling period of 30 minutes and two 30 minutes counting intervals after sampling. At a flow rate of 22.5ℓ/min and a detector efficiency of 17% the sensitivity for each individual daughter concentration is better than 0.03 pCi/ℓ based on 20% of statistical accuracy.

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This system is well suited for continuous operation, too. However, radon- and thoron-daughters can only be determined in terms of potential alpha energy concentrations.

INTRODUCTION

In order to obtain detailed information about the levels of radon, thoron and their daughters indoors, sensitive equipment is needed. Since outdoor concentrations play an important role in well-ventilated houses the methods should be capable of detecting variations of atmospheric levels with satisfying accuracy.

For this purpose existing methods, e.g. scintillation flasks, ionization chambers or two-filter devices for radon or thoron detection are not sensitive enough or the equipment is too bulky to be used in homes.

Adequate methods to measure radon daughters using the filter technique may be satisfying for grab sampling of radon daughters only. Since a considerable contribution of the potential alpha energy concentration derives from thoron daughters, too (WICKE and PORSTENDORFER, 1980) only such methods can be used which are able to discriminate between both radon and thoron daughters.

Many of those naturally occurring radionuclides are alpha emitters with almost different alpha energies, hence alpha spectroscopic methods are advantageous to use. The type of detector which can easily be applied for this purpose is the surface-barrier detector, a semiconductor.

In our studies specific instrumentation has been developed which uses those detectors for measuring low levels of radon, thoron and their daughters in air.

Radon and Thoron Measurement

The method we applied for radon and thoron detection is a specific version of the two-filter technique. Radon and thoron gas enter a decay volume while their daughters are trapped by an entrance filter. Instead of collecting the daughters which are produced during the passage of the gas on an exit filter, initially positively charged Po-281 (RaA)- and Po-216 (ThA)- atoms are removed by a strong electric field. The daughters, collected directly on the aluminized surface of a surface-barrier detector can easily be identified by alpha spectroscopy. Under adequate conditions it can be shown that the decay rate of Po-218 and Po-216 is proportional to the radon concentration and the thoron concentration, respectively.

Such type of instrumentation was presented by JACOBI (1963) with a volume of 140ℓ, capable of detecting concentrations of radon as low as 0.1 pCi/ℓ within an hour. Similar devices with smaller volumes and scintillation detectors were used by ROBERTS AND DAVIES (1966) and DALU and DALU (1971) for the detection of atmospheric concentrations of radon.

Since those instruments, in contrast to the two-filter technique, do not need high flowrates, diffusion of radon gas was sometimes sufficient. Those "passive" instruments were proposed for the application in mines (COSTA RIBEIRO et. al., 1969) and in homes (WRENN, SPITZ and COHEN, 1975).

The type of instrument we are using consists of a 14.2ℓ aluminium sphere, which incorporates a surface-barrier detector* isolated in a PVC-mounting (Fig. 1). The entering air is dehumidified by Ca Cl_2, and radon- and thoron-daughters are removed by an entrance filter. For grab sampling the sphere can be evacuated and the air be sucked in at the place of interest. After applying the high-voltage counting can start initially.

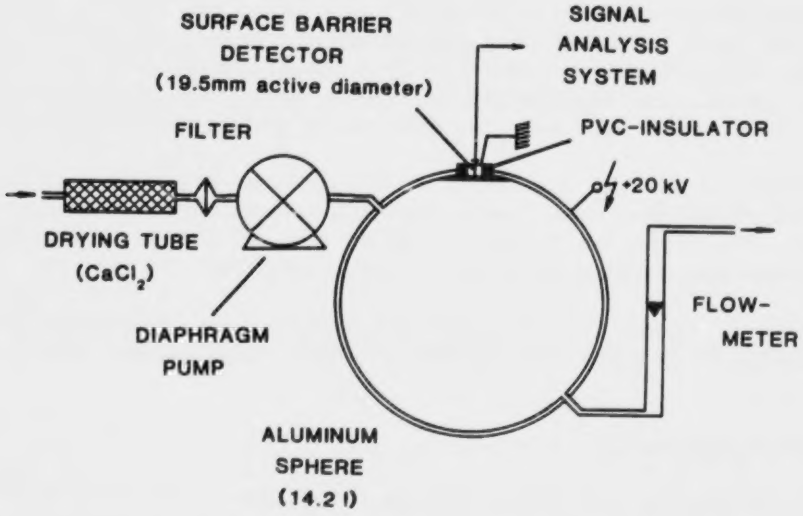

Fig. 1: Electrostatic collection chamber system for radon and thoron measurements

For field purposes we combined the detector with a "state-of-the-art" preamplifier, shaping amplifier and a portable multichannel analyzer with a fast tape recorder** (Fig. 2).

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* Ortec CR-24-300-100, 19.5mm active diameter.

**Silena NIM Standard Multichannel Analyser.

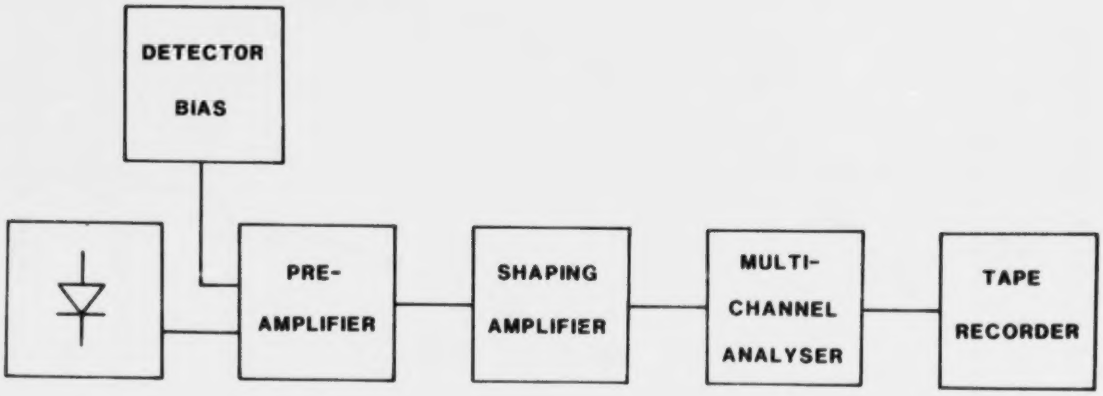

Fig. 2: Alpha particle signal analysis system

Since radon and thoron daughters are deposited directly on the sensitive surface of the detector, a very good energy resolution is obtained (~80KeV). Fig. 3 shows a typical alpha spectrum of a radon-thoron gas analyzed by this technique.

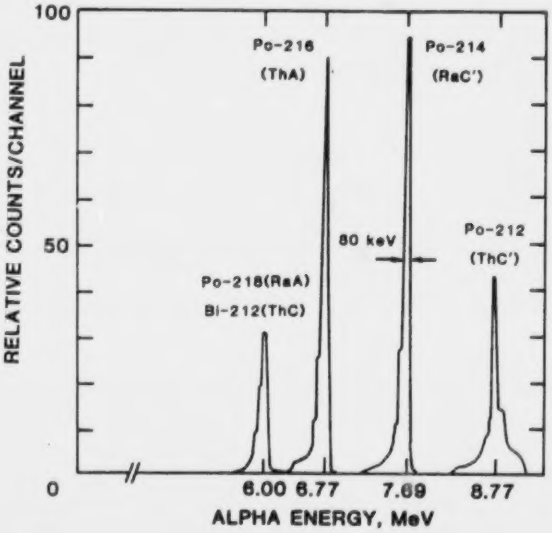

Fig. 3: Alpha spectrum of a radon/thoron daughter mixture resulting from electrodeposition on a surface-barrier detector

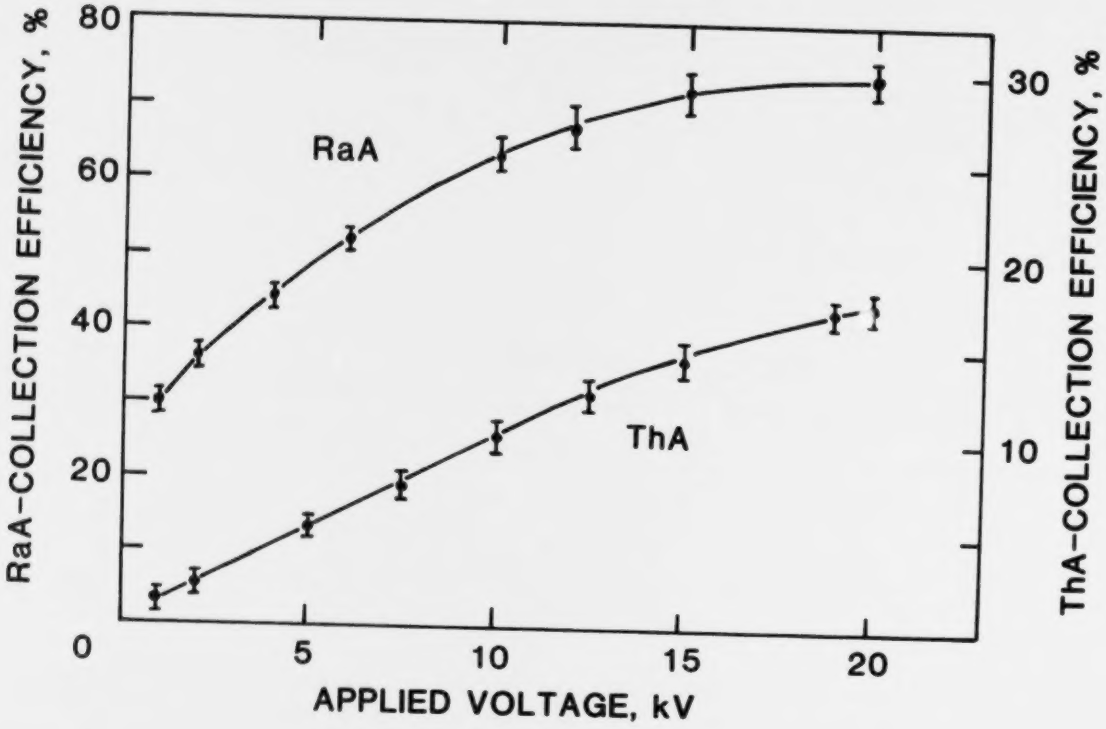

Fig. 4: Collection efficiency for Po-218 (RaA) and Po-216 (ThA) as a function of the applied voltage

In order to obtain a high-efficient electrodeposition the strength of the electric field is very important. According to Fig. 4 saturation for Po-218 collection is achieved above 16kV, while the collection rate for Po-216 is still increasing.

For practical purposes we applied 20kV to the system and obtained a satisfactory collection rate.

A critical factor is the influence of humidity. Fig. 5 shows that between 0 and 50% relative humidity, small changes have a considerable influence on the collection efficiency of Po-218. Best results are obtained when the air is carefully dried.

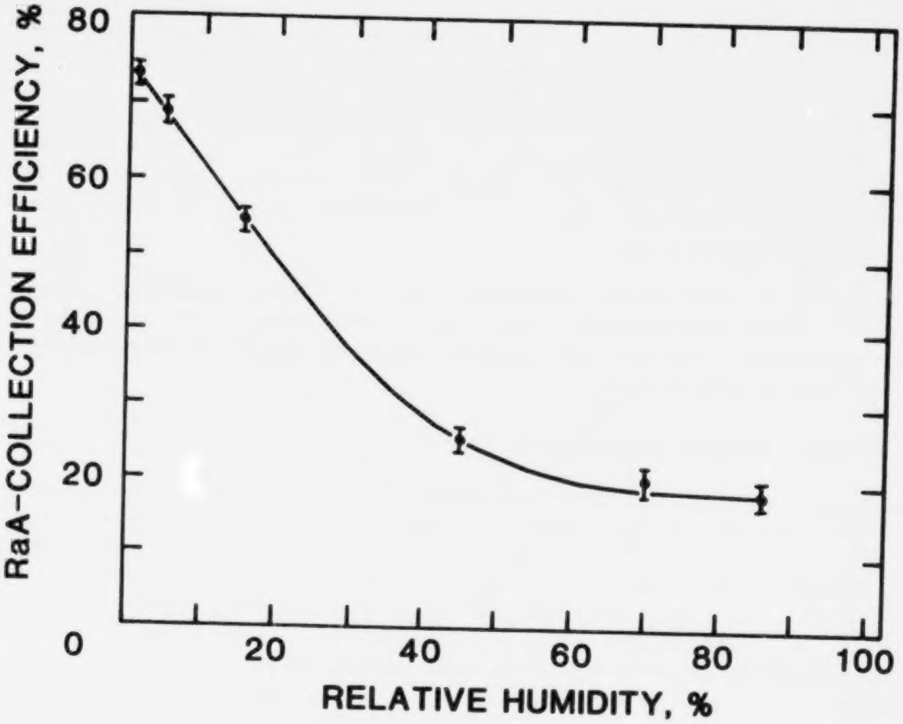

Fig. 5: Collection efficiency of Po-218 (RaA) as a function of relative humidity

The calibration of the system was done by introducing a known amount of radon into the sphere emanating a standard radium solution. The average value of several calibrations was 12.0 cpm(pCi/ℓ). At a one-hour counting time the sensitivity for radon is 0.03 pCi/ℓ assuming an error of 20% to counting statistics.

The detection efficiency for thoron of the static version of the instrument is so poor (~4 counts/(pCi/ℓ) for counting times > 10 minutes) that only the flow-through type of this instrument is of interest. For moving the air a small diaphragm pump* and a flowmeter are added to the instrument (see Fig. 1).

A flowrate of 1.6ℓ/min results thoron detection efficiency of about 0.4 cpm/(pCi/ℓ) and a sensitivity of 1 pCi/ℓ for a one-hour counting interval. These values improve for higher flow rates. For radon, however, the detection efficiency is fairly independent of the flow rate, 12 cpm/(pCi/ℓ).

We applied this instrument successfully in those studies, where quick changes of the radon concentration should be monitored. At a flow rate of 1.6ℓ/min the response time of the system was only about 10 minutes depending on the size of the drying tube.

Radon- and thoron-daughter measurement

An established method for the determination of radon-daughters in air is to draw a certain volume of air through a membrane or a glass-fiber filter and to count the alpha activity on the filter. The Tsivoglou-Thomas method (THOMAS 1972) uses gross alpha counting in three time intervals after collection to calculate the RaA, RaB and RaC-concentration in air. The accuracy, especially for RaA, was improved by CLIFF (1978) taking the first counting period during sampling. Although more sophisticated instrumentation is necessary, an alpha spectroscopic analysis fo the filter sample can improve accuracy. Such a procedure was proposed by MARTZ and co-workers (1969) and presented for practical applications in a recent paper by NAZAROFF et.al. (1981). Making a similar approach as CLIFF, DUGGAN AND HOWELL showed how to improve the spectroscopic technique by taking the first counting while sampling was in progress.

Adopting this method for our purposes radon- and thoron-daughters were collected on a membrane filter during a sampling period of t_o, while alpha-spectroscopic counting of Po-218 (RaA) and Po-214 (RaC') took place (t_1-t_2) using a surface-barrier detector. During a second time interval from t_3-t_4 only those counts from Po-214 were taken. Once those three alpha counts are obtained it is easy to calculate the individual radon daughter concentrations by the method of "Simultaneous Equations" (WICKE 1981).

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*Wisa type 203

To optimize the precision of the technique one must carefully select the sampling and counting intervals. The calculations show that the precision of Po-218 measurement can be improved by longer sampling/counting intervals, if the assumption of a constant concentration in air would be applicable. The precision of Pb-214 and Bi-214 (Po-214)* measurement is associated with the time lag between the end of sampling and the start of the second counting interval. Table 1 displays the selected sampling and counting periods applied for this method with associated sensitivities. Here the sensitivity is defined as the concentration in air of a radon daughter such that the standard deviation in the measurement, associated only with the random nature of radioactive decay, is 20% of the sampled concentration. For comparison purposes the product of flow rate and detector efficiency is assumed to be equal to 1ℓ/min and the activity ratio of the daughters Po-218:Pb-214:Bi-214 = 1:0.6:0.4. Comparing the sensitivities of this alpha-spectroscopic technique with other methods (Table 1) it is evident that the procedure presented here can be used to make radon daughter measurement more precisely.

As an extension of this method counts of Po-212 at 8.77 MeV at two different counting intervals after sampling can successfully be used for the determination of thoron daughter concentration in air. In order to obtain a good sensitivity for both Pb-212 and Bi-212 specific sampling and counting intervals have been selected (Table 1). The sensitivities, due to 20% statistical error, which are presented in this table are based on an assumed activity ratio Pb-212:Bi-212= 1:0.5.

To obtain a good energy resolution of the alpha-spectroscopic instrument as well as a good detection efficiency a sampling unit has been designed consisting of a surface-barrier detector[1] in a distance of 6mm from a membrane filter[2] (Fig. 6). The air is drawn through this filter by a diaphragm pump[3] at a relative constant flow rate of 22.5ℓ/min. The signal analysis system is similar to the one used for the radon/thoron monitor (Fig. 2).

The detection efficiency of the system can easily be calculated using a Monte-Carlo method (WILLIAMS 1966) and be checked by a standard plane source. For the geometry of our system a detector efficiency of 17% ± 0.5% was obtained.

The energy resolution of this alpha-spectroscopic device is good enough for a satisfactory separation of Po-218 (6.00MeV), Po-214 (7.69MeV) and Po-212 (8.77MeV) with an overlap significantly below 1% (Fig. 7).

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*Po-214 is assumed to be an initial radioactive equilibrium with Bi-214.

1: Ortec type CR-24-300-100, 19.5mm active diameter
2: Sartorius membrane filter type SM, 1.2mm pore size, 25mm diameter, 20mm effective diameter
3: Neuberger type NO35.1.2.AN18

TABLE 1: COMPARISON OF METHODS FOR MEASURING RADON DAUGHTER CONCENTRATIONS

METHOD/REFERENCE	SAMPLING AND COUNTING TIMES, MIN				20%-SENSITIVITY*, pCi/ℓ		
	T_0	T_1- T_2	T_3- T_4	T_5- T_6	Po-218	Pb-214	Bi-214
THOMAS (1972)	5	7-10	11-25	26-35	37.0	5.9	13.4
CLIFF (1978)	10	0-10	11-26	27-42	3.7	1.6	2.5
	15	0-15	16-36	37-57	2.4	0.5	1.2
KEEFE ET.AL (1978)	3	3.2-6.2	-	-	** 2.9	4.2	2.6
NAZAROFF ET.AL. (1981)	5	6-15	24-40	-	1.4	1.7	1.0
	10	11-21	31-50	-	1.1	0.7	0.6
THIS WORK	10	0-10	20-50	-	0.4	0.4	0.4
	20	0-20	40-60	-	0.2	0.2	0.2
	30	0-30	60-90	-	0.1	0.1	0.1
					Pb-212	Bi-212	
THIS WORK	20	20-40	60-80	-	0.35	0.18	
	20	20-40	100-120	-	0.13	0.2	
	30	30-60	60-90	-	0.27	0.20	
	30	30-60	90-120	-	0.13	0.10	

* STATISTICAL ERRORS CONSIDERED ONLY; EFFICIENCY = 100%; FLOWRATE = 1ℓ/MIN;
EQUILIBRIUM RATIO: Po-218:Pb-214:Bi-214 = 1:0.6:0.4, Pb-212:Bi-212 = 1:0.5

** DATA ADOPTED FROM NAZAROFF ET.AL. (1981)

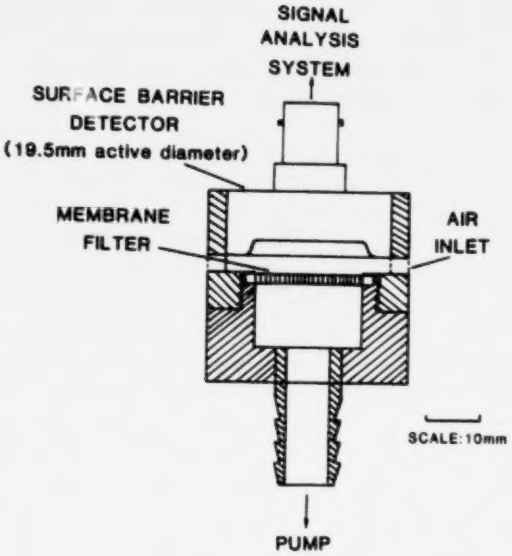

Fig. 6: Sampling unit for a simultaneous collection and detection of alpha emitters in air

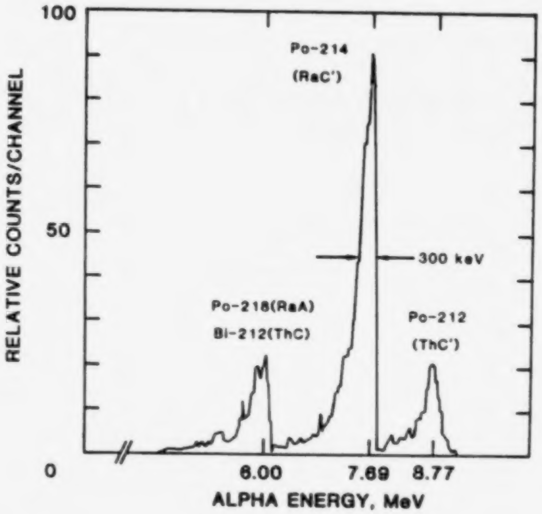

Fig. 7: Alpha spectrum obtained by collecting airborne radon- and thoron-daughters on a membrane filter.

At a flow rate of 22.5ℓ/min and a detector efficiency of 17% the sensitivity for each individual daughter concentration is better than 0.03 pCi/ℓ based on the data of Table 1 for 30 minutes of sampling and counting periods of 3 x 30 minutes after sampling.

Continuous operation of the system is possible. Since sampling takes place all the time information is available only about the current alpha activity of Po-218 (RaA), Po-214 (RaC') and Po-212 (ThC') on the filter. This is not sufficient to determine all 5 individual radon- and thoron-daughter concentrations. But it is possible to calculate the potential alpha energy concentration for each time interval from the integrated counts (HAIDER and JACOBI 1974, KAWAJI 1981). The advantage of this instrument is its capability to distinguish between the potential energy concentrations originating from radon- and thoron-daughters. Work on this subject is still in progress; details will be presented later.

CONCLUSION

Alpha-spectroscopic methods using surface-barrier detectors are well suited for the analysis of radon, thoron and their daughters in air. It has been demonstrated that a specific design of instrumentation leads to a high sensitivity for grab-sampling application as well as to good time-response at continuous operation.

ACKNOWLEDGEMENTS

This work was financially supported by the Federal Ministry of the Interior, Federal Republic of Germany.

Participation at this conference was made possible by support of the New Mexico Institute of Mining and Technology, Socorro, New Mexico.

References

CLIFF, K.D. (1978): The measurement of low concentrations of radon-222 daughters in air, with emphasis on RaA assessment. Phys. Med. Biol. 23, 55-65.

COSTA-RIBEIRO, C., THOMAS, J., DREW, R.T., WRENN, M.E., EISENBUD, M. (1969): A radon detector suitable for personal or area monitoring. Health Phys. 17, 193-198.

DALU, G., DALU, G.A. (1971): An automatic counter for direct measurement of radon concentration. J. Aerosol Sci. 2, 247-255.

DUGGAN, M.J., HOWELL, D.M. (1968): A method for measuring the concentration of the short-lived daughter products of radon-222 in the atmosphere. Int. J. Appl. Radiat. Isot. 19, 865-870.

HAIDER, B., JACOBI, W. (1974): A new monitor for long-term measurement of radon daughter activity in mines. P. 217-226 in: Radiation Protection in Mining and Milling of Uranium and Thorium. Proceedings of a conference in Bordeaux, France, 9-11 September 1974.

JACOBI, W. (1963): A new method to measure radon and thoron in flowing gases and its use to determine the thoron content of atmospheric air. Colloque International sur la Pollution Radioactive des Milieux Gaseux, Saclay, France.

KAWAJI, J., LIN PAI, H., PHILLIPS, C.R. (1981): Use of gross filters activities in a continuous working level monitor. Health Phys. 40, 543-548.

KEEFE, D.J., McDOWELL, W.P., GROER, P.G. (1978): The environmental working level monitor. Report No. P7628C, Argonne National Laboratory, Argonne, Illinois.

MARTZ, D.E., HOLLEMAN, D.F., McCURDY, D.E., SCHIAGER, K. (1969): Analysis of atmospheric concentrations of RaA, RaB and RaC by alpha-spectroscopy. Health Phys. 17, 131-138.

NAZAROFF, W.W., NERO, A.V., REZRAN, K.L., (1981): Alpha-spectroscopic technique for field measurement of radon daughters. Paper presented at the Second Special Symposium on Natural Radiation Environment, Bombay.

ROBERTS, P.B., DAVIES, B.L. (1966): A transistorized radon measuring equipment. J. Sci. Instr. 43, 32-35.

THOMAS, J.W. (1972): Measurement of radon daughters in air. Health Phys. 23, 783-789.

WICKE, A. (1981): Comparison of different methods for radon daughter measurement. Laboratory Report New Mexico Institute of Mining and Technology, Socorro, New Mexico.

WICKE, A., PORSTENDORFER, J. (1980): Exposure to radon daughters in dwellings. Paper presented at the Specialist Meeting on the Assessment of Radon and Daughter Exposure and Related Biological Effects, Rome, March 3-7, 1980.

WILLIAMS, I.R. (1966): Monte-Carlo calculation of source-to-detector geometry. Nucl. Instr. Meth. 44, 160-162.

WRENN, M.E., SPITZ, H., COHEN, N. (1975): Design of continuous digital output environmental radon monitor. IEEE Trans. Nucl. Sci. NS-22, 645-648.

RECENT ADVANCES IN THE DESIGN AND USE OF PASSIVE RADON MONITORS

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ABSTRACT

A passive environmental radon monitor (PRM) based on the electrostatic collection of ^{218}Po ions on a thermoluminescent dosimeter (TLD) was introduced by Andreas George (1977). Since then, the device has undergone considerable testing, modification and field use by several developers.

The important changes to the passive environmental radon monitor have been increased volume, a more sensitive TLD, convenient dessicant cartridges, a safe and reliable case, an improved TLD holder, and a non-changeable dessicant replacement.

This paper describes AeroVironment's experience with the development and use of the PRM and the present state of the art.

INTRODUCTION

In 1978, AeroVironment Inc. performed environmental monitoring in the vicinity of proposed uranium mines in north-central Wyoming. One of our tasks was to collect ambient radon concentration data.

Following accepted procedures, we used Tedlar bags and small, regulated air pumps to collect representative 48-hour samples. We had the usual problems associated with this method. The air bags leaked. The pumps quit. On occasion snow and mud prevented us from reaching the sampling locations at the appointed times.

These problems in monitoring radon led us to search for a better method. We found the answer in a passive radon monitor, described by George and Breslin (1979) and Cowper and Davenport (1978). Using their description as a starting point, an equipment manufacturer built the FR-3 for us.

We began field testing several of the passive radon monitors (PRMs) early in 1979, comparing results with the Tedlar bag method. The results of this effort were presented to the IEEE (Friedland et al., 1980) and eventually referenced by the revised version of NRC Ref. Guide 4.14. As Tables 1 and 2 show, we found our PRMs results essentially comparable to the Tedlar bag results, without the problems.

TABLE 1. Comparison results of Tedlar bag method vs. PRM method.

Dates	Tedlar Method (PCi/l)	PRM Method (pCi/l)
4/30/79-5/8/79	0.79 ± 0.05	1.03 ± 0.08
5/1/79-5/9/79	0.43 ± 0.04	0.52 ± 0.04
4/30/79-5/8/79	0.43 ± 0.04	1.68 ± 0.05
5/1/79-5/9/79	0.65 ± 0.04	1.70 ± 0.06
6/19/79-6/25/79	0.62 ± 0.05	0.42 ± 0.03
6/4/79-6/12/79	1.07 ± 0.06	0.81 ± 0.06
6/5/79-6/13/79	0.69 ± 0.05	0.68 ± 0.05

TABLE 2. Averaged comparison results of Tedlar bag method vs. PRM method.

	Tedlar Method (PCi/l)	PRM Method (pCi/l)
Combined average over a season at a given site	0.75 ± 0.06	0.69 ± 0.06
	0.52 ± 0.04	0.47 ± 0.03
	0.75 ± 0.05	0.74 ± 0.05
	0.66 ± 0.04	0.69 ± 0.05

CONSTRUCTION AND THEORY OF PRM

The PRM is usually comprised of a three-compartment cylinder. The principle component of the unit is the ionization chamber, with its thermoluminescent dosimeter (TLD)-bearing cathode. Accessory to this are a small bottom chamber, which contains a layer of dessicant sandwiched between two wire grids with filters; and a top chamber, which contains the battery supply for maintaining an electric field between the cathode and the walls of the ionization chamber. The entire PRM is encased in a cover of PVC or metal. The battery and ionization chambers are separated by a solid plate, which incorporates a central opening for the cathode. The top of the battery compartment is protected by a lid.

The primary TLD, which is covered by an aluminized mylar window, is positioned on the tip of a cathode, which protrudes downward into the ionization chamber. Radon diffuses through the filters and dessicant and into the bottom of this chamber. Radon-daughters are removed by the filters. As the radon in the chamber undergoes radioactive decay, each emitted alpha particle is likely to induce a loss of more than two electrons. Hence, the resultant radium-A ions generally are positively charged and tend to be deposited on the negatively charged window of the cathode. As each deposited radium-A ion decays, the energy from the emitted radiation has a definable probability of being trapped by the TLD. This also holds true for successive members of the decay chain. Most of the energy deposited in the TLD is from the alpha particles which have been emitted by radium-A and radium-C'. However, due to the possibility of significant TLD exposures by cosmic and ambient terrestrial radiations, background TLDs may prove necessary. These are placed on the cathode but are shielded from alpha radiation.

Our first PRM, the FR-3, has the basic construction we have described but it differs from later models in that it is all metal. Other distinguishing features include short metal legs and a battery supply consisting of two 510-volt cells. While we have fairly good results with this unit, we were not entirely happy with it. The battery arrangement did not allow good wiring. Inserting the cathode between the batteries could result in an electric shock. Inconveniently, the dessicant had to be poured out a small window for drying. The LiF TLDS we used at first were not as sensitive as we thought they should be.

MODIFICATIONS TO PRM

Intending to correct the problems noted above, we asked our supplier for a new model, dubbed the FR-32. It was constructed mostly of PVC material. Graphite powder was used to coat the inside of the ionization chamber. The dessicant cartridge was easily removable, allowing the TLD probe to be inserted from the bottom. Four batteries of 300 volts each were used in place of the previous battery supply. The more sensitive TLD-200's (CaF_2-Dy) were used as the dosimeters. However, we still had a few problems. The graphite lining of the ionization chambers proved unsatisfactory and was replaced by a metal liner. The battery compartment lids often leaked and allowed battery damage. The dessicant cartridges were made of PVC and hence could not withstand the high temperatures required for speedy dessicant drying. This was corrected in the later model of the FR-32 substituting a metal dessicant cartridge.

Our latest PRM, called the LR-5 (diagrammed in Figure 1), has no dessicant cartridge at all. Instead we have a thin sheet of material at the bottom which allows radon to diffuse in, but keeps the moisture out. The new battery compartment lid is a press-fit PVC cap. This never has to be removed, except for the infrequent battery changes. A small PVC pipe passes through the lid and battery compartment. The TLD probe is inserted into this small pipe where it makes electrical contact. The portion of the probe containing the TLD protrudes down into the ionization chamber. The new probe also eliminates some of the TLD breakage we had experienced earlier.

Thus, our present PRM is the result of considerable development and refinement. However, nearly all the modifications were performed with an aim of greater safety, convenience and dependability. The size and shape of the ionization chamber and the positioning of the TLD have changed very little in the development of our PRM. The 8-liter, eight-inch diameter ionization chamber has been standard. In each PRM, the active TLD is covered by a mylar window and positioned 1-3/4 inches from the top of the ionization chamber. Following early testing with a 1/8" LiF TLD, the current 1/8" x 1/8" x 0.015" CaF_2-Dy TLD was chosen and has been the exclusive TLD in our monitoring. Since the TLD and ionization chamber constitute the heart of the system, we have had a consistent frame of reference for comparison and calibration purposes.

Performance of the LR-5 and its predecessor, the FR-32, were compared in March and April, 1981. Table 3 shows the results, which indicate that, indeed, the two are comparable.

CALIBRATION

The calibration of the PRMs involves two separate processes. First, the TLDs are calibrated using gamma source at a known distance according to NRC and ANSI standards. A secondary standard is used to check TLD performance. Then, semi-annually, individual PRMs are exposed to known radon concentrations and calibrated from the TLD readouts of these PRMs. Several of these calibrations have been performed, including both laboratory and field exposures. The results are fairly comparable, as evident in Table 4.

For TLD readout, we have used both the Harshaw Model 2000 A & B and the Teledyne 7300. We have been pleased with the performance of each of these units. It should be noted that each TLD reader has its own characteristic response, which will be different from that of other TLD readers, even with identical settings on units of the same model number. Hence, new calibrations are required with each reader.

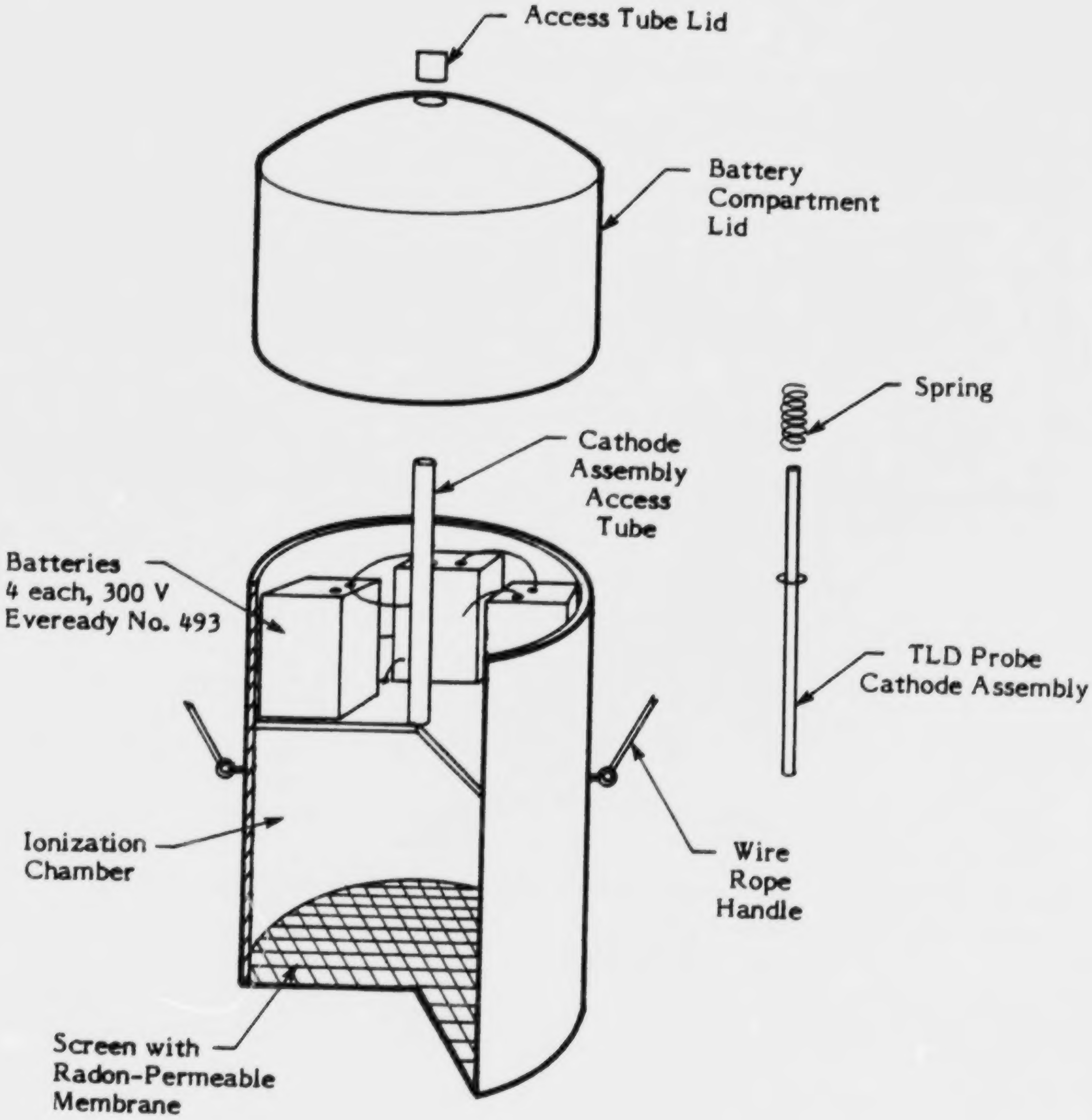
Access Tube Lid
Battery Compartment Lid
Spring
Cathode Assembly Access Tube
Batteries 4 each, 300 V Eveready No. 493
TLD Probe Cathode Assembly
Ionization Chamber
Wire Rope Handle
Screen with Radon-Permeable Membrane

FIGURE 1. Schematic diagram of the LR-5 Passive Radon Monitor.

TABLE 3. Field intercomparison of different PRM models.

		FR-32 (pCi/l)	LR-5 (pCi/l)
Site 1	March 81	0.33	0.35
	April 81	0.39	0.38
Site 2	March 81	0.28	0.27
	April 81	0.25	0.31
Site 3	March 81	0.40	0.32
	April 81	0.37	0.29

Table 4. PRM calibration results.

PRM Model No.	Calibration Facility	Test Period	Total Radon Exposure (pCi/l)hr	Calculated Calibration Factor $\frac{\text{nc}}{\text{pCi/l hr}}$
FR-3	New Mexico Institute of Mining & Technology	Nov 79	79.9	0.0601
FR-3	Denver Mining Research Center	Apr 80	48,700	0.0411
FR-32	New Mexico Field Calibration	Dec 80	88.8	0.0590
FR-32	New Mexico Field Calibration	Jul 81	412.9	0.0597
LR-5	Wyoming Field Calibration	Jul 81	415.5	0.0588

CONCLUSIONS

In conclusion, it should be noted the LR-5 Passive Radon Monitor is the end product of considerable testing and modification. It is safer and more reliable than earlier models, yet it maintains the desired level of durability. It also retains the capability to monitor normal ambient concentrations.

ACKNOWLEDGEMENTS

We would like to thank the following individuals for their help and advice:

Of the Denver Mining Research Center of the U.S. Bureau of Mines -- Robert Droullard, Ted Davis, Emery Smith and Robert Holub.

Of the Environmental Measurements Laboratory of the U.S. Department of Energy -- Al Breslin, John Harley, and Andreas George.

References

George, A.C., and A.J. Breslin (1979): Measurements of environmental radon with integrating instruments. Health and Safety Laboratory, U.S. Energy Research and Development Laboratory (now EML, DOE) New York, NY 10014. Published in the Workshop on Methods for Measuring Radiation in and Around Uranium Mills, by the Atomic Industrial Forum, Inc., Washington, D.C. 20014.

Friedland, S.F., L. Rathbun and H.M. Goldstein (1980): Radon monitoring: uranium mill field experience with a passive detector. Paper presented at the Nuclear Science Symposium of the IEEE, October 17-19, 1979, San Francisco, CA.

Friedland, S.F., L. Rathbun and J. Smith (1979): Radon monitoring near a uranium mill with a passive detector. Paper presented at the 2nd Symposium on Uranium Mill Tailings Management. Colorado State University, Fort Collins, CO., Nov. 19-20.

Cowper, G., and M.R. Davenport (1978): An instrument for the measurement of long-term average radon levels. Chalk River Nuclear Laboratories, Ontario, Canada. Published in International Atomic Energy Agency, publication number SM-229/100.

TRACK ETCH ®RADON DETECTOR CALIBRATIONS AND FIELD RESULTS

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ABSTRACT

Initial calibration results with the Track Etch radon detection system have been previously reported.(1) Additional calibration data have recently been obtained in a wide variety of standard radon test chambers and preliminary results from this work can be reported. Seven different radon test facilities have been utilized for these tests in addition to the DOE Environmental Measurements Laboratory which was utilized for the initial tests. Six different detector configurations have been calibrated including two new miniaturized configurations which may be suitable for personnel dosimeters as well as other applications. Individual calibration tests have used 2 to 20 replicates of each detector configuration. Radon exposures have ranged from 20 to 10,000 (pCi/l)-days at radon concentrations from 5 to 1000 pCi/l. Results give mean values for each set of replicates as a function of the total exposure. Calibration factors and detector sensitivities derived from the tests will be discussed.

Large numbers of Track Etch detectors have been and are being used for radon monitoring in homes, buildings and outdoors. The results from some of these measurements will also be discussed.

INTRODUCTION

The Track Etch ® radon measuring technique has been widely used in a passive manner to make time integrated radon concentration measurements.(2) Applications of this technique have included radon soil gas measurements for uranium exploration, earthquake prediction, geothermal exploration, and fundamental geological and geophysical studies.(3,4) Track Etch detectors are now being extensively used for monitoring radon in houses, buildings, uranium mines and processing facilities and in other areas where there is a need to know exact radon concentrations.(5,6)

With the Track Etch technique, an alpha sensitive plastic detector (usually mounted in a plastic container or on a card) is exposed to the atmosphere to be measured. Alpha particles from radon in the air, or from radon daughters plated out on various adjacent surfaces, penetrate the detector and cause radiation damage tracks that are subsequently revealed by an etching process.

The number of alpha tracks counted per unit area (T/sq.mm) is therefore proportional to the average exposure (eg. (pCi/l)-hours). Exposure times can range up to a year or more, if desired, using improved detectors which retain alpha tracks without fading for very long time periods.

Early types of Track Etch detectors suffered from deficiencies in sensitivity, track fading and optical quality, although early tests showed the promise of a passive integrating device. In recent years, an improved type of detector has been developed which overcomes the deficiencies of the earlier type. This detector has already seen wide use in a number of radon measuring applications during the last three years.

Track Etch detectors have several features that make them particularly attractive for environmental radon monitoring uses. They are simple, small and lightweight, and they have no moving parts, electronics or power supplies. They are also sensitive only to alpha particles and are not sensitive to any gamma or beta radiation that may be present. Since the detectors are so small and have no moving parts, they can be easily used for making unobtrusive radon measurements in occupied houses and buildings. The etched detectors also provide a permanent record for future reference.

TRACK ETCH DETECTOR CONFIGURATIONS

Several different Track Etch detector configurations have been developed for different applications. Either radon-only or combined radon and radon daughter measurements can be made. The various configurations are described below:

Open Cup (Type C). The Type C Track Etch detector system consists of a small plastic cup with the detector element attached to the inside bottom of the cup. This open cup system is the one usually used in making soil gas measurements for mineral exploration and earthquake prediction applications. In the Type C configuration, the detector measures the primary radon gas alpha particles originating within the cup as well as the alpha particles from the radon daughters plated-out on the inside walls of the cup. The Track Etch reading for an open cup is not only a function of the radon and radon daughter activities, but it is also a function of the plate-out characteristics of the ambient atmosphere. It is therefore not recommended for other than soil gas measurements. (Figure 1)

Membrane Covered Cup (Type M). The Type M Track Etch detector system is the same as the Type C except that the opening of the cup is covered with a semi-permeable plastic membrane.(7,8) This membrane (sometimes called a Thoron Filter) slows the normal diffusion of noble gases into the cup and thus discriminates against radon-220 (sometimes called thoron that has a 55 second half-life) while permitting 60 to 70% of radon-222 (which has a 3.8 day half-life) to enter the cup. The membrane covered cup system is mainly used in making soil gas measurements for mineral exploration applications to eliminate thoron interference and water accumulation in the cup. It also prevents the entrance of radon daughters and thus measures a radon-only signal.

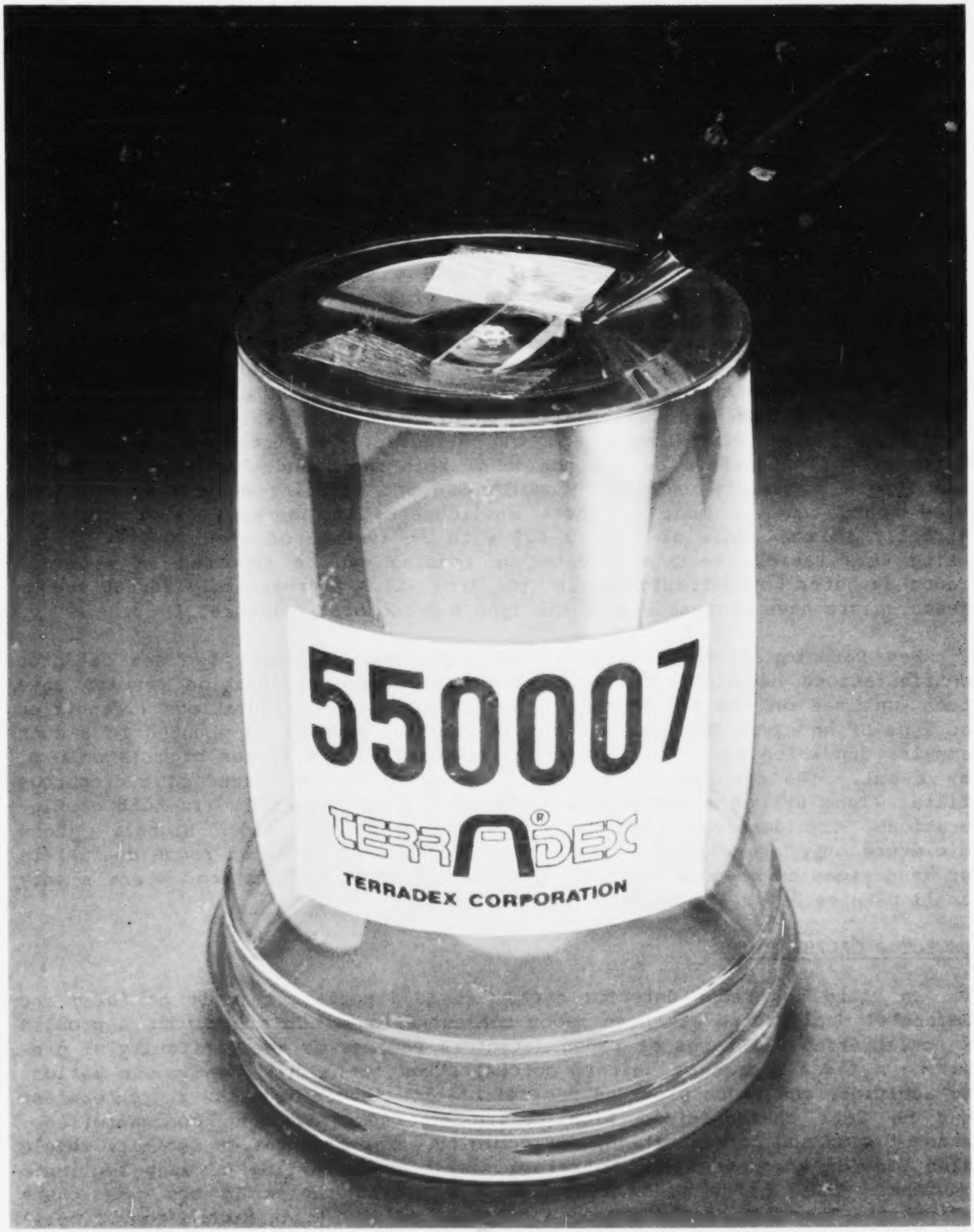

Figure 1. Type-C Open Cup Track Etch Detector

Filter Covered Cup (Type F). The Type F Track Etch detector system is the same as the Type C except that the opening of the cup is covered with a hydrophobic, microporous filter which permits rapid and complete infiltration of the gaseous radon isotopes but discriminates against the nongaseous radon daughters. Because of its higher sensitivity to radon, the Type F configuration is preferred to the Type M configuration in all radon-only applications where thoron is not an important component.

Bare Detector Card (Type B). In this configuration the Track Etch detector element is mounted flat on a card facing the atmosphere to be measured. The effective measured volume is a hemisphere with a radius of 3.0 inches (7.68 centimeters), the range of the polonium-212 alpha particle in air, or 2.5 inches (6.30 centimeters), the range of polonium-214 alpha particle if only the radon-222 series is present. In using the Type B card detector, it is mounted in such a way that it is always at least 4 inches (10 centimeters) from any adjacent surface to eliminate any alpha contribution from plated-out radon daughters on that surface. The bare detector measures radon plus ambient radon daughters and is sensitive to plate-out only with respect to radon daughters plated out directly on the detector. The reading on the bare detector will therefore be a function of both radon and the radon daughters. The Type B is mainly used for indoor house and building monitoring. It has been found that in most of these environments the Working Level Ratio is normally in the range of 0.3 to 0.7 with an average of about 0.5(9,10) By using this ratio, the Type B detector reading can be reported in radon or radon daughter concentration units (pCi/l or WL). Approximately 26,000 indoor measurements have been made with the Type B detectors. (Figure 2)

New Detector Systems (Type SF and Type SM). Recently two new detector configurations have been developed and preliminary calibration results have been obtained on the two systems. In the two new configurations (identified as Type SF and Type SM), the Track Etch detector element is mounted in a much smaller cup which is 1 1/4 inches in diameter and 3/4 inches high (3 cm O.D. by 2 cm). The opening of the cup is covered by the standard microporous filter (Type SF) or the standard membrane filter (Type SM). In both configurations, the detector element responds only to the radon signal. These miniature cups are expected to be ideal for use as personal radon dosimeters or as devices to measure radon in homes or in other applications where a very small passive device is needed. (Figure 3)

DETECTOR CALIBRATIONS

In using any radon detector system it is important that it be fully and accurately calibrated at known radon concentrations. This can pose a problem in calibrating all types of radon detectors because of the difficulty of providing a radon source of uniform concentration for a long continuous period. In addition, the radon provided for calibration should have low thoron content and be well characterized with regard to radon daughter concentrations, aerosol size and concentration, and humidity. The calibration facility should also provide adequate space to permit simultaneous testing of many replicate devices. Only a few radon test chambers around the world can meet these criteria. The test chambers chosen for most of the Track Etch detector calibrations reported here were those operated by the U.S. Department of Energy

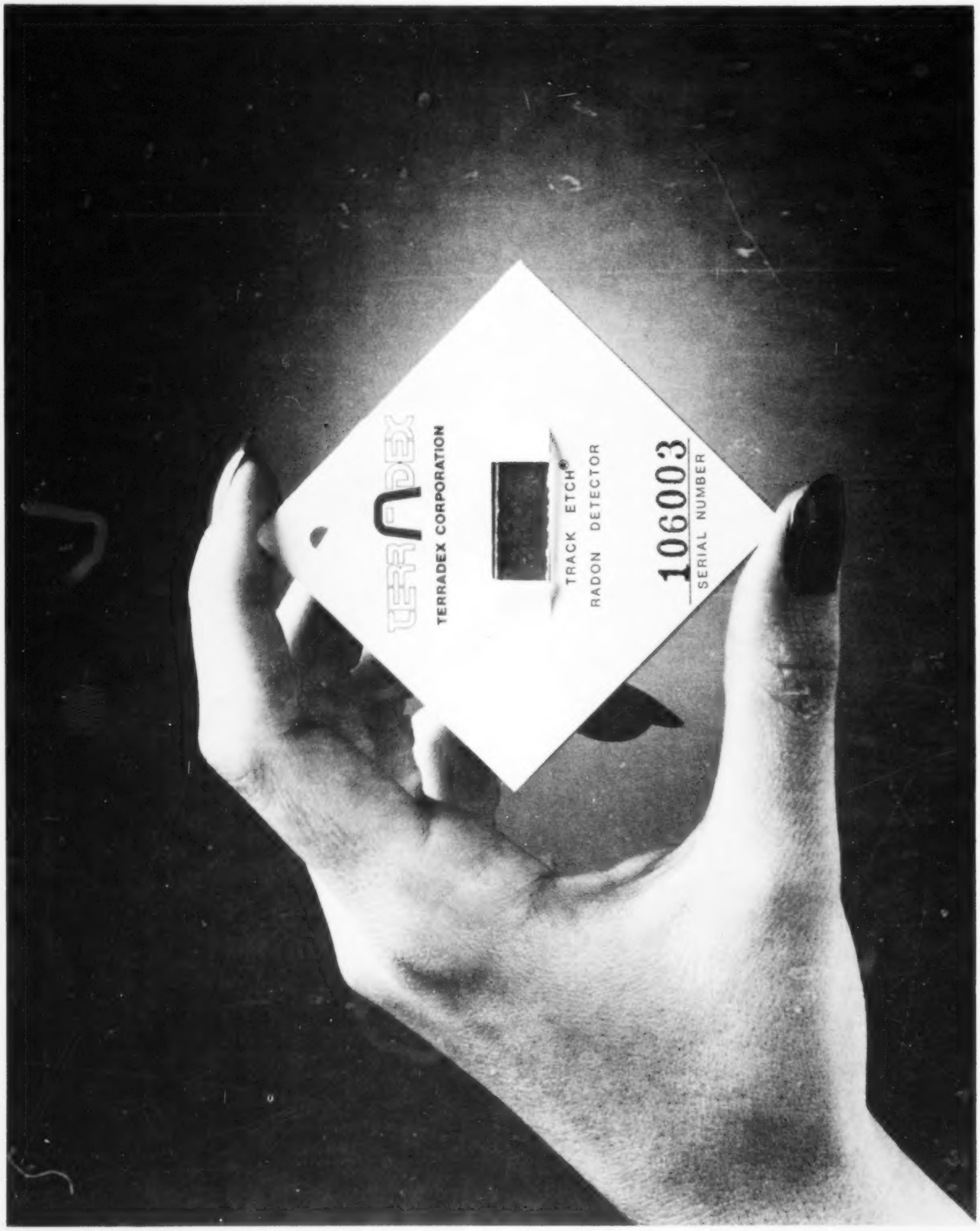

Figure 2. Type-B Bare Detector Card Track Etch Detector

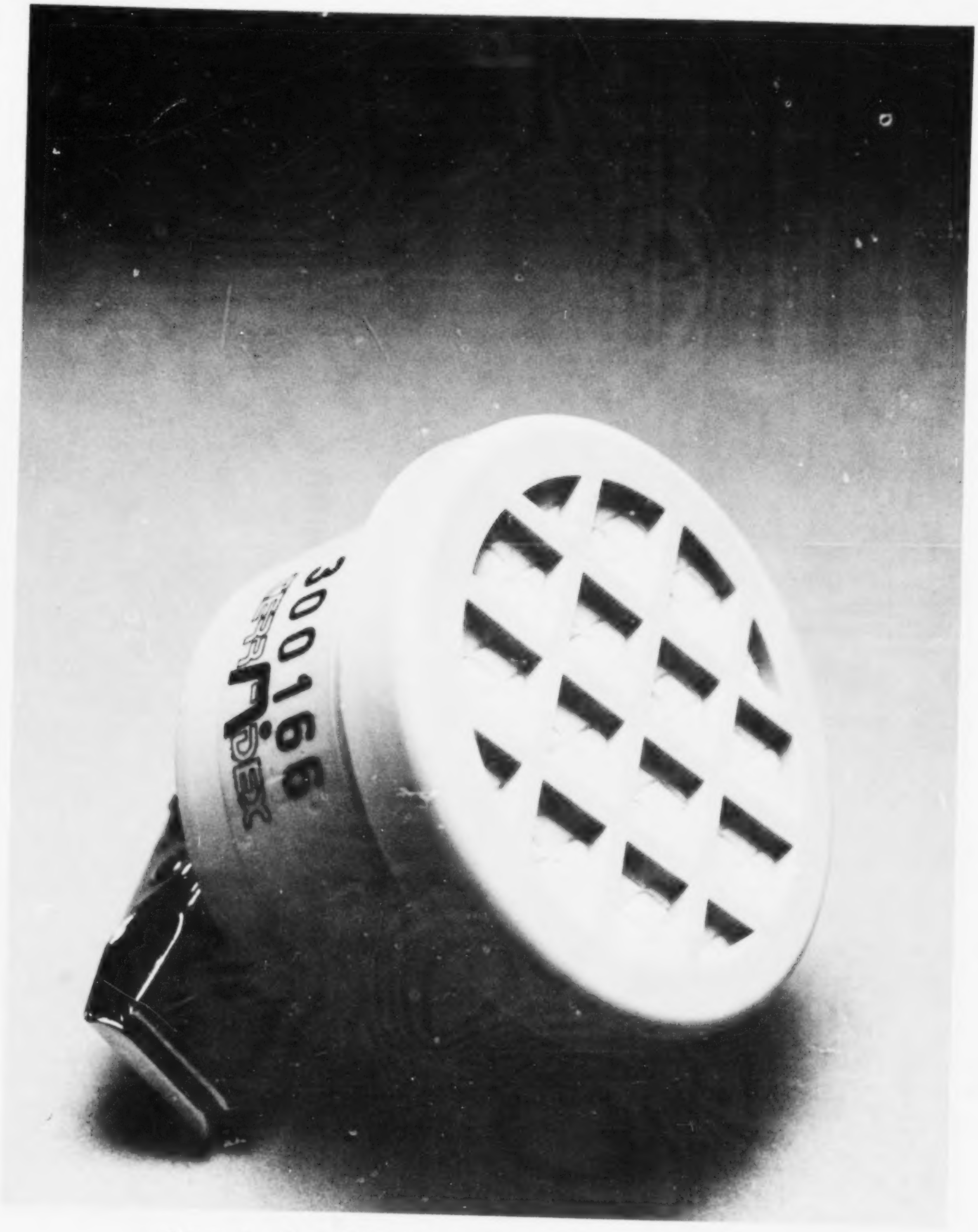

Figure 3. Type-SF Track Etch Personnel Radon Dosimeter

at the Environmental Measurements Laboratory (EML), although we have subsequently calibrated the detectors in four other test chambers. The earliest EML calibration tests were conducted in a small (2 cubic meter) test chamber while a larger test chamber (20 cubic meters) was extensively used in the later phases of the calibrations. Radon concentrations in both test chambers were measured using a continuous scintillation flask monitoring system. The Working Level Ratios approximated the ratios found in most indoor environments (0.3 to 0.7).

The test exposure conditions for the calibrations in the EML are shown in Table 1. Each of four different detector types was calibrated and in some instances as many as 20 replicate samples were used for each detector type. After the detectors were exposed, they were returned to Terradex Corporation for processing and reading. At least 100 tracks were counted for each detector to keep the relative standard deviation to less than 10% due to counting statistics. The relative standard deviation measured from each set of replicates agreed with counting statistics.

Table 2 combines the Track Etch detector readings with the radon exposures as reported by the Environmental Measurements Laboratory. Each detector reading is the mean of the replicate readings. To obtain the appropriate calibration factors, the track density readings were divided by the measured radon exposure values and the resulting data are presented in Table 3. Table 3 also shows, for each configuration, the mean value and related statistics. The mean and its standard deviation are calculated by weighting the Table 3 values by the number of replicates at each exposure. The error includes any error in the radon exposure as measured by the chamber operator. The higher standard deviations of the Type C and Type B configurations reflect the varying radon daughter concentration at each exposure.

An example of the replicate detector response from a single radon test exposure is shown in Table 4. These data show that there is excellent sample to sample reproducibility of the four different detector types. The percent uncertainty (relative standard deviation) of a single detector is in the range of 7 to 13%.(1) This is consistent with the expected uncertainty due to counting statistics for a single detector.

Since this calibration work was completed, several other calibration tests have been made, both at EML and in other chambers in 1980 and 1981, for the Type F and Type B configurations. Additional calibration and intercomparison work is continuing at six different radon test chambers on a regular basis.

Mean calibration factors for all 1979 and 1980 runs fall within 1% of the 1979 FILT value reported in Table 3 and within 10% of the Type B value. Mean T/sq.mm values for all runs are plotted against (pCi/l)- days in Figs. 4 and 5 together with the weighted mean lines from Table 3. These figures show the data from all of the EML tests as well as that obtained from three other test chambers.(5)

TABLE 1

EML RADON TEST CHAMBER EXPOSURE CONDITIONS

Calibration Test	Chamber	Duration (Days)	Average Radon Exposure Rate (pCi/l)	Detectors Calibrated
1	$2m^3$	14	1085	5
2	$20m^3$	120	38	3
3	$2m^3$	21	350	15
4	$20m^3$	79	25.6	20
5	$20m^3$	36	11.6	20
6	$20m^3$	115	21.2	20
7	$20m^3$	30	33.2	80
8	$20m^3$	93	32.6	80
9	$20m^3$	63	32.1	80
			Total -	323

TABLE 2

RADON EXPOSURE VS. DETECTOR READING

Calibration Test	Duration (Days)	Radon Exposure ((pCi/l)-days)	Detector Reading (Mean Tracks/sq.mm)			
			Type C	Type M	Type F	Type B
1	14	15190	-	-	-	1608
2	120	4660	-	-	-	638
3	21	7350	556	259	-	851
4	79	2020	90.6	31.9	55.6	115.1
5	36	418	25.7	9.7	17.0	23.5
6	115	2440	125.0	41.8	78.2	137.2
7	30	996	45.2	19.2	34.2	73.2
8	93	3030	200.9	73.8	126.5	297.6
9	63	2020	110.9	45.8	77.5	206.0

TABLE 3

CALIBRATION FACTORS				
Calibration Test	(Tracks/sq.mm)/(pCi/l)-day			
	Type C	Type M	Type F	Type B
1	-	-	-	.1068
2	-	-	-	.1369
3	.0756	.0352	-	.1158
4	.0448	.0158	.0275	.0570
5	.0615	.0232	.0407	.0562
6	.0512	.0171	.0320	.0562
7	.0454	.0193	.0343	.0735
8	.0663	.0244	.0417	.0982
9	.0549	.0227	.0384	.1020
Weighted Mean	.0562	.0223	.0372	.0891
Relative Standard Deviation of the Mean (%)	2.2	2.5	1.8	2.8

TABLE 4

REPLICATE READINGS FROM TWENTY DETECTOR SAMPLES

Detector Number	Detector Reading (Tracks/sq.mm)			
	Type C	Type M	Type F	Type B
1	49.6	20.5	28.8	73.3
2	50.0	18.7	34.4	80.3
3	50.0	15.9	39.6	70.5
4	47.9	18.4	36.8	66.5
5	29.5	17.0	28.9	71.9
6	35.1	18.6	30.0	71.9
7	44.4	19.3	35.1	78.9
8	50.5	15.4	31.2	67.2
9	46.6	18.2	35.8	73.3
10	43.5	20.5	30.3	74.0
11	40.4	20.3	36.5	74.0
12	47.4	20.7	33.0	82.5
13	57.2	21.0	29.5	77.5
14	45.7	19.0	37.5	73.3
15	46.6	21.2	38.9	64.5
16	41.3	19.5	34.4	77.9
17	46.1	19.3	32.2	78.2
18	43.5	21.6	38.9	78.2
19	43.9	19.7	34.7	67.2
20	45.7	19.9	36.5	64.0
Mean	45.2	19.2	34.2	73.2
Std. Dev.	5.8	1.7	3.5	5.4
Rel.Std.Dev.(%)	13	9	10	7

Note: Test conducted in 20 cubic meter EML test chamber at radon exposures of 996 (pCi/l)-days

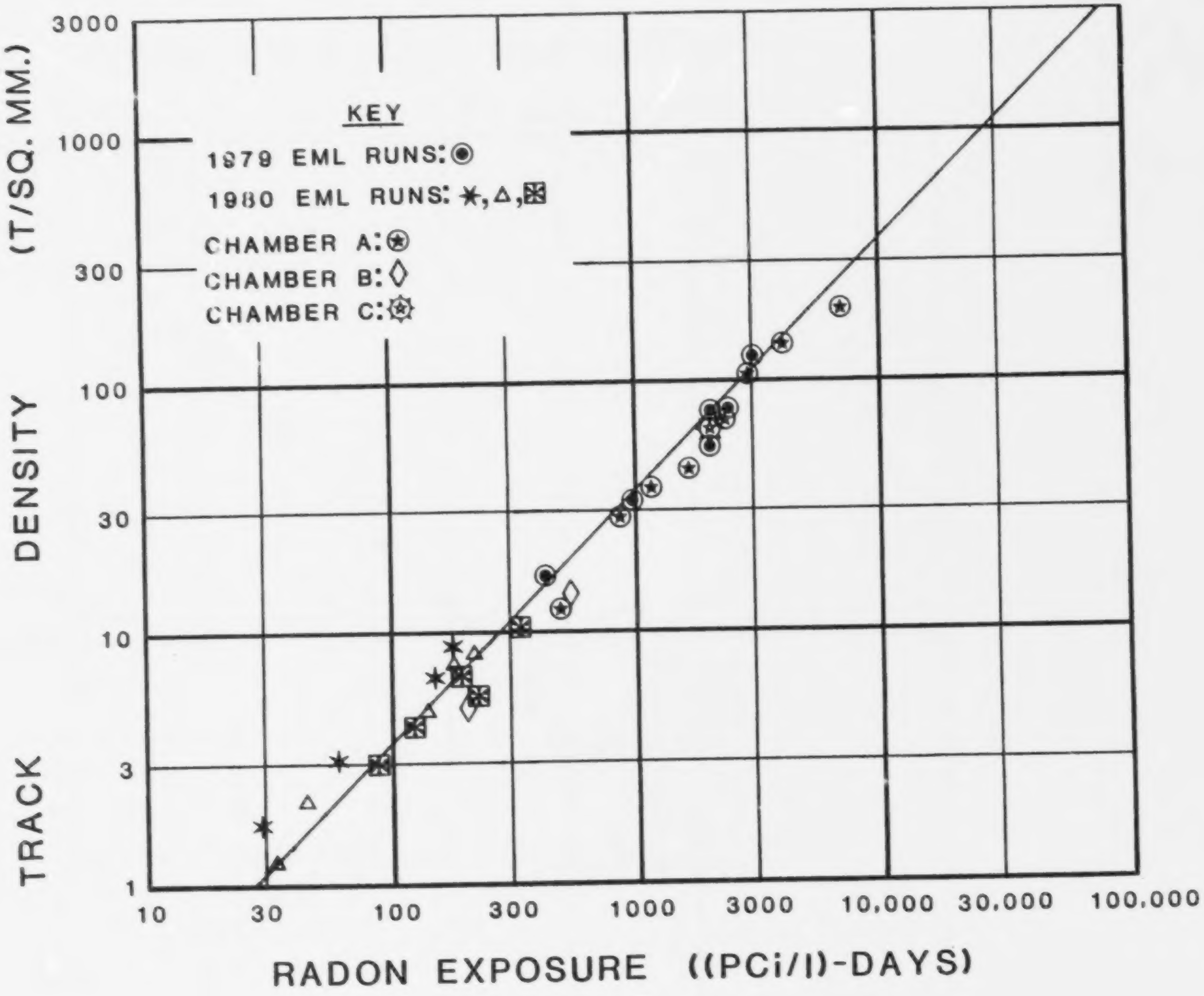

Figure 4. Type-F (Filtered Cup) Detector Calibration

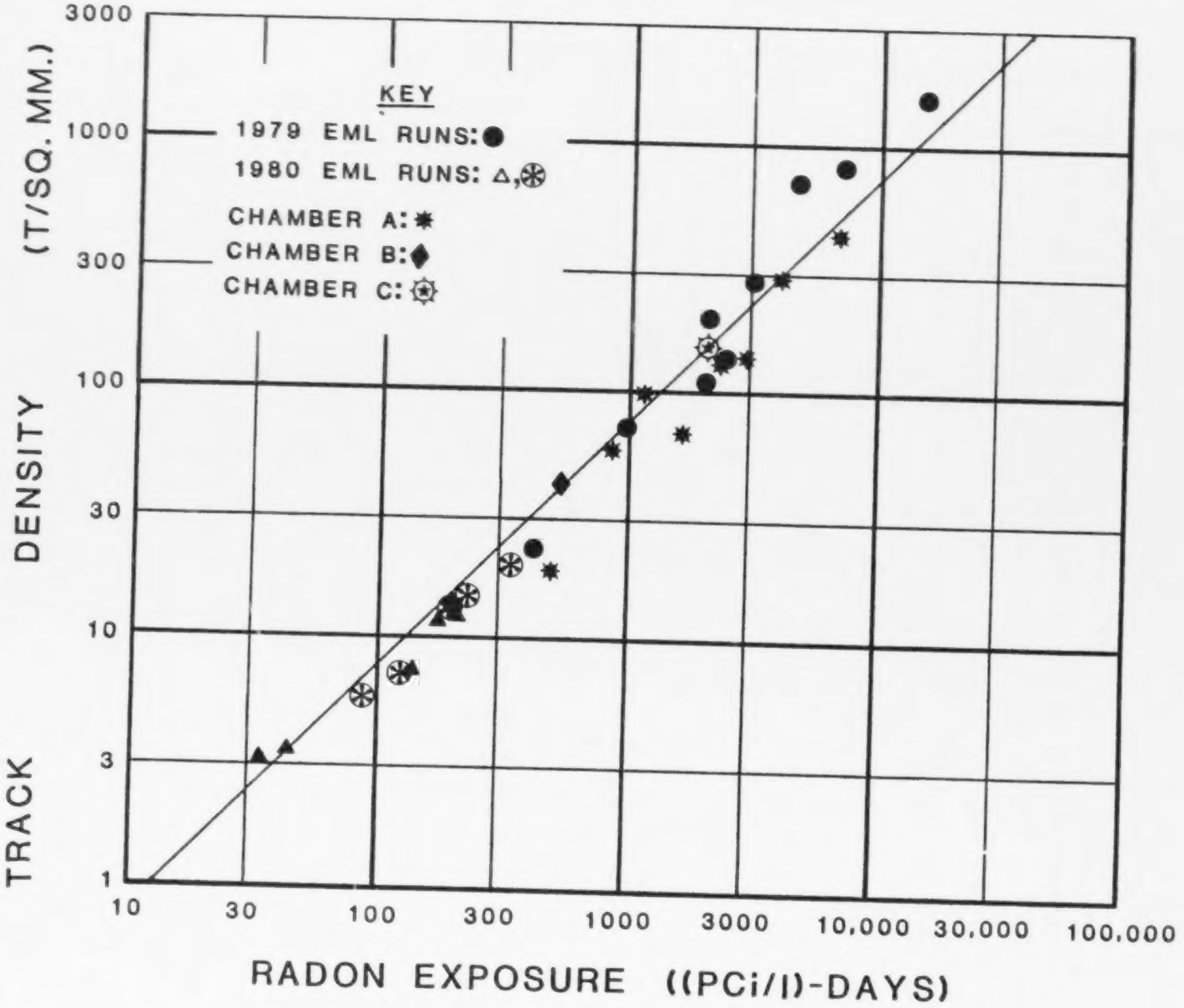

Figure 5. Type-B (Bare Card) Detector Calibration

The plots show the somewhat increased (over counting statistics) scatter of the data for the Type B configuration which is responding to the variable daughter and plate-out environment in each calibration test.

Using the statistical information from the calibration tests, the sensitivity of the different detector configurations was calculated as a function of the area counted on each detector.(5) The results show that a one to three month integrating period is adequate for a variety of outdoor or indoor measurements at typical radon concentrations (See Table 5).

TABLE 5

TRACK ETCH DETECTOR SENSITIVITY*

DETECTOR AREA COUNTED (SQ.MM)	RADON EXPOSURE ((pCi/l)-Months)			
	Type C	Type M	Type F	Type B
1.15	2.6	6.5	3.8	2.0
5.75	0.7	1.8	1.0	0.5
17.25	0.4	0.8	0.4	0.2

*Sensitivity is that measured radon exposure whose relative standard deviation due to all sources of error is 50%.

FIELD RESULTS

At the present time, Track Etch detectors are being used to make measurements in a variety of environmental monitoring situations. The largest use is for house and building monitoring, mostly in selected areas where high radon concentrations are expected. Other uses include monitoring around operating uranium mines, mine tailings and mill sites, baseline studies around planned uranium mill sites, monitoring around phosphate and radium processing sites, monitoring in radium needle storage facilities and measuring radon in soil gases.

Outdoor Surveys

To make outdoor radon measurements, the Type F detector configuration is usually employed, and it is often placed in a canister to protect it from the weather elements. The Track Etch detectors are ideally suited for outdoor measurements since they can make long time-integrated readings and they require no power supplies or maintenance of any kind. These features are of particular value in the outdoor environment, such as near uranium mining and milling facilities where the radon levels vary greatly as a function of production activity, weather and season, and where a temperature-insensitive passive device is essential. (Figure 6)

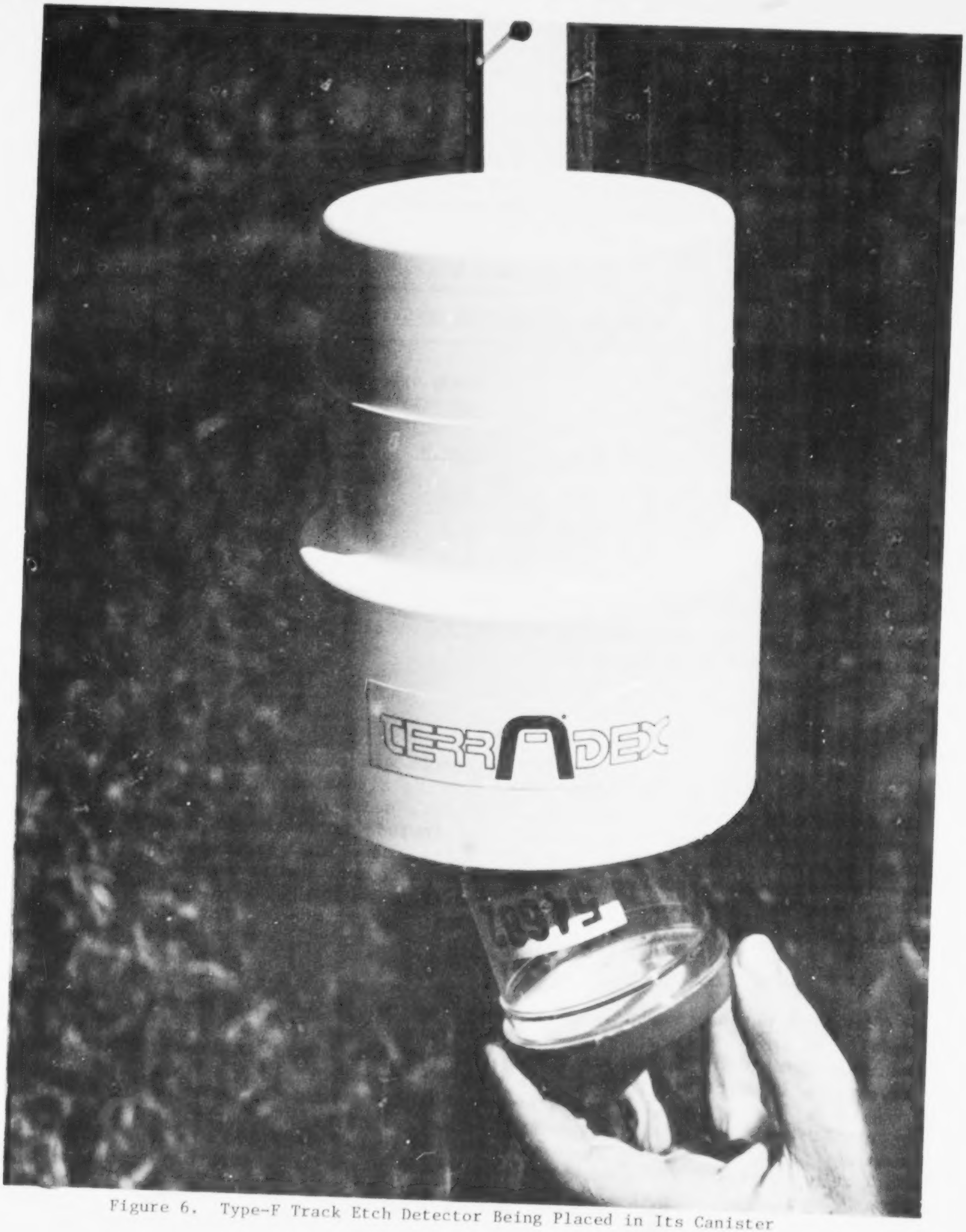

Figure 6. Type-F Track Etch Detector Being Placed in Its Canister

Preliminary results from the radon measurements around one operating uranium mine and mill complex have been previously reported.(6) In this measurement program, 12 sampling stations were established around the facilities and the detectors were left in place for a month at a time. This program has now been continued for a full year and the data from three typical sampling stations are shown in Fig. 7. At each station it can be noted that there is a fairly large month to month variation depending on the activities at the facility and the changing weather patterns. The maximum variations are typically two to three times the average concentrations, and they vary by factors of 5 to 10 from the highest months to the lowest months at the same locations. Normally the on-site measurements were significantly higher than either the site boundary or the off-site measurement. The on-site measurements for the year averaged 2.62 pCi/l while the site boundary averaged 1.18 pCi/l and the off-site averaged 0.89 pCi/l. It is interesting to note, however, that during the month of May the off-site location had a slightly higher average radon concentration than the other two sites. The year-long average radon concentrations show a typical fall-off with distance from the center of the mine/mill complex. (Figure 8)

Indoor Surveys

To make indoor measurements, either the Type B detectors or the Type F detectors are usually employed. Most indoor measurements made to date have been made with the Type B detector cards, since they are easier to handle and use in occupied dwellings. (The difference in response of these two detector types was discussed previously.) The detector cards are usually attached to the wall or hung from the ceiling of the rooms being monitored. Often two or more detectors are placed in each home being monitored, and the sampling locations are selected on the basis of expected occupancy rate and those locations suspected of having high radon concentrations.

The statistical data from almost 30,000 indoor Track Etch radon measurements in seven different major locations are shown in Table 6. The locations include: Northern California; Houston, Texas; Northeastern U.S.; Eastern Pennsylvania; the State of Maine; Saskatchewan, Canada, and numerous locations in Sweden. Many of these measurements were made in ordinary homes not suspected of having high radon concentrations, but many (particularly those in Sweden) were made in homes suspected of having higher than normal radon concentrations due to location or due to building materials. Most of the houses were built with the normal construction practices for that general location and few were known to be especially constructed for energy efficiency. In most instances the Track Etch detectors were left in place for 3 months but individual detectors were left in place as short as 1 month or as long as 10 months. The summertime periods were usually avoided when making the measurements because of the tendency to have unusually high ventilation rates at these times due to warmer weather.

The general statistical results from the indoor surveys are shown in Table 7. In Table 7, radon concentration data are given in picoCuries per liter (pCi/l) averaged over the exposure period. Statistics show the mean radon concentration (which is close to the median value in all surveys), the low and high values, and the percent of the data exceeding 20 pCi/l. Table 7 also shows these results converted to Working Level (WL) assuming a Working Level Ratio (WLR) of 0.5. The 20 pCi/l level (0.1 WL) would expose an individual to about 2 Working Level Months (WLM) per year.

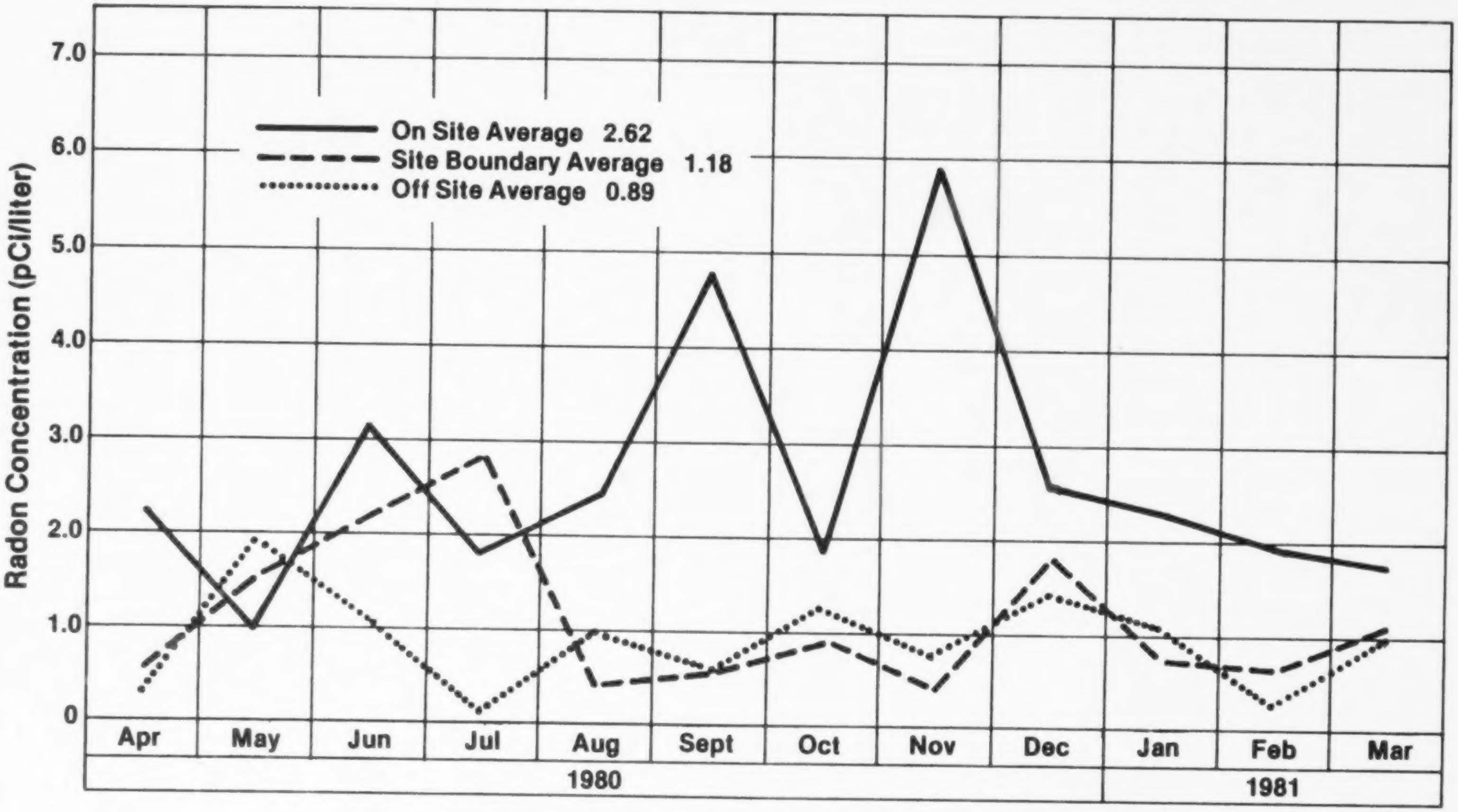

Figure 7. Atmospheric Radon Around a Uranium Mine-Mill Complex

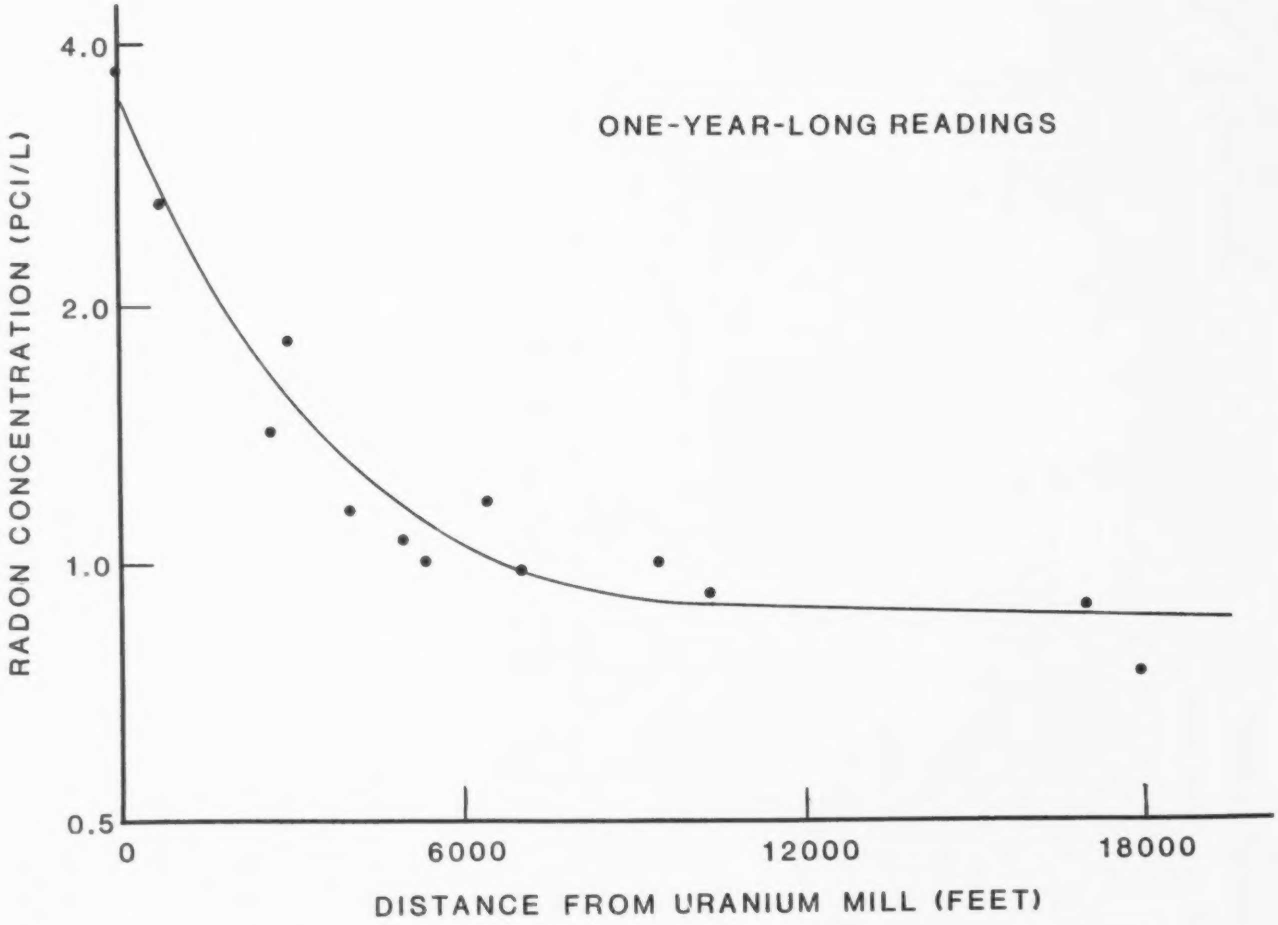

Figure 8. Reductions in Radon Concentrations Away From a Uranium Mill

TABLE 6

SUMMARY OF INDOOR TRACK ETCH MEASUREMENTS

Location-Season	Detectors	Integrating Time (Mos.)
N. Calif. winter-summer	30	3
Houston fall-winter	304	2-5
No. East U.S. summer	51	3
E. Penna. winter-spring	249	2-8
Maine fall-spring	427	3-10
Canada winter-spring(1)	124	3
Sweden winter-summer	112	1-3
Sweden spring-summer(1)	750	1-4
Sweden fall-winter(1)	4709	1-4
Sweden winter(1)	9397	1-4
Sweden winter-spring(1)	13164	1-4
Total	29317	

(1) Selected Homes

TABLE 7

INDOOR TRACK ETCH RESULTS*

Location-Season	pCi/l		Working Level		Percent Greater Than 20PCi/l
	Mean	Range	Mean	Range	
N. Calif. winter-summer	0.5	0.1-2.1	0.002	0.0005-0.01	0
Houston Fall-winter	0.4	0.1-2.7	0.002	0.0005-0.01	0
No. East U.S. summer	0.3	0.1-3.6	0.002	0.0005-0.02	0
E. Penna. winter	2.4	0.1- 91	0.01	0.0005-0.46	15
Maine fall-spring	1.1	0.1-133	0.006	0.0005-0.66	3
Canada winter-spring	3.6	0.1- 34	0.02	0.0005-0.17	4
Sweden winter-summer	2.1	0.4- 22	0.01	0.002 -0.11	4
Sweden spring-summer	4.1	0.2-106	0.02	0.001 -0.53	4
Sweden fall-winter	8.5	0.2-520	0.04	0.001 -2.6	14
Sweden winter	10.0	0.5-466	0.05	0.002 -2.3	15
Sweden winter-spring	9.5	0.5-1140	0.05	0.002 -1.8	12

*Expressed in Average Radon or Radon Daughter Concentrations and Assuming Working Level Ratio of 0.5

The radon concentrations in those homes from Northern California, Houston and Northeastern U.S. in the summertime were relatively low and they clearly show the influence of warm weather and increased ventilation. The high concentrations from Eastern Pennsylvania and Maine show the dual effects of reduced air change rates and high radon production rates from the uranium containing soils and rocks under and around these homes.

In Sweden, the radon concentrations in ordinary houses are higher than the low U.S. results, undoubtedly due to tight Swedish construction practice. (Selected Swedish homes are from communities pinpointed by geology and construction materials which are thought to lead to high radon values.) The Canadian homes were selected from a group showing 0.01 WL or greater by previous grab sampling.

A significant number of the homes measured in Eastern Pennsylvania and Sweden have radon concentrations exceeding 20 pCi/l (0.1 WL). This is a concentration at which there may be a significant concern about an increase in lifetime lung cancer risk. The high concentrations were somewhat expected in the Swedish locations but were unexpected in many of the other locations. The highest concentrations measured in a number of homes are at levels unacceptable for workers in uranium mines. It should also be noted that present indoor provisional standards in the 0.02 WL (4 pCi/l) range may be considerably below the actual levels in many normal homes.

Figure 9 shows the statistical distribution of the indoor radon concentrations from 4709 measurements from Sweden and from all of the measurements made in Maine and Houston.(11) The data from these and all other indoor surveys have a log-normal distribution. In addition, the data show the range of values that can be expected as they are influenced by geology and building ventilation. Both the Swedish and Maine results are high because of geology, construction materials and the lower air change rates in cold climates. The Houston data represent well-ventilated homes in a warm climate with apparently little uranium mineralization in the underlying soil and rocks.

CONCLUSIONS

The results from calibration tests and field measurements demonstrate that the Track Etch technique for radon monitoring provides adequate sensitivity and reproducibility for both indoor and outdoor measurements. The technique is also practical and easy to use when making large-scale, long-term radon measurements. The completely passive nature of the detectors makes the Track Etch technique particularly attractive for making measurements in any locations where it is desirable that they be left unattended for a period of time. The use of selected types of Track Etch detector systems permits the selective measurement of radon only and the elimination of thoron response.

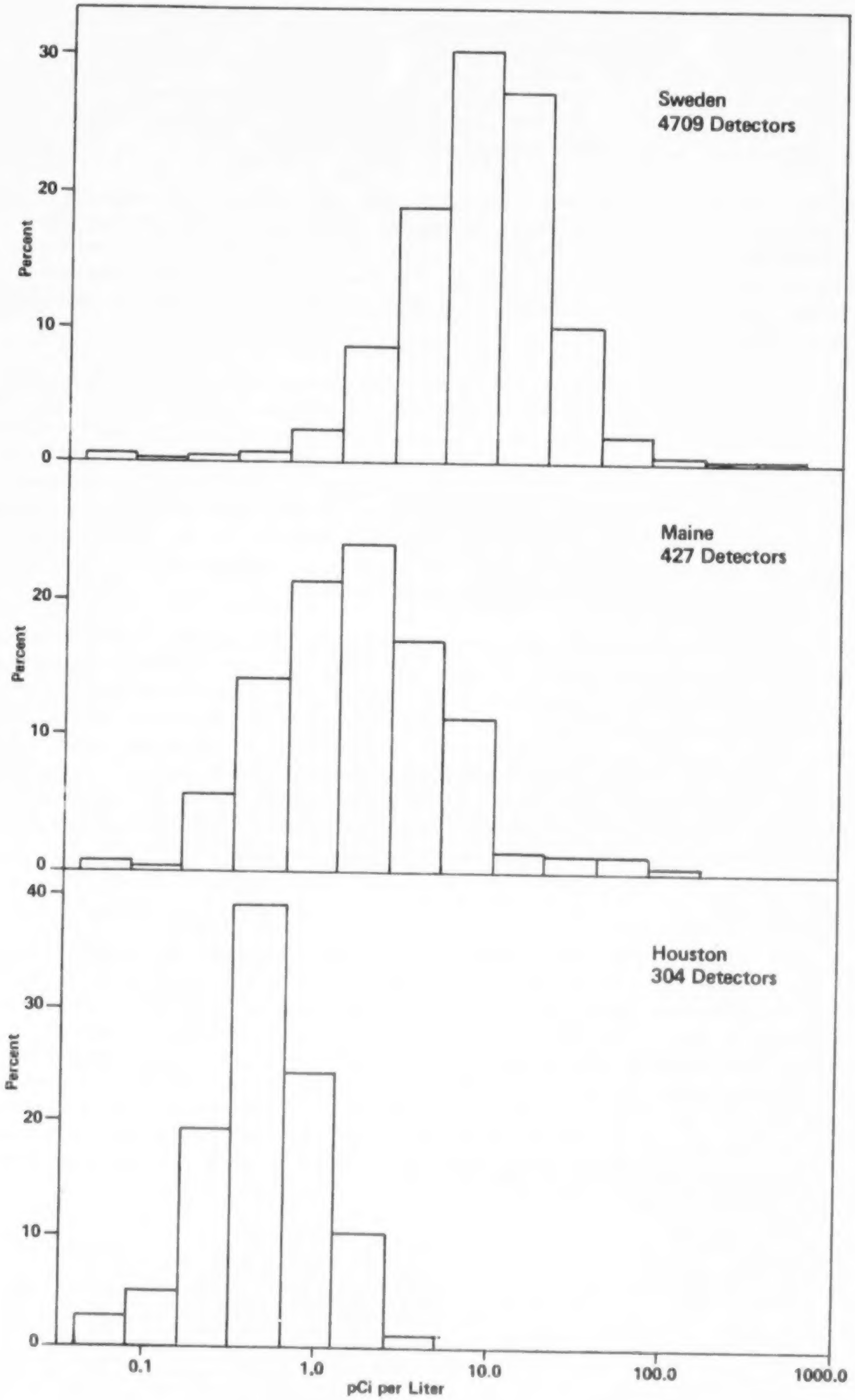

Figure 9. Statistical Distribution of Indoor Radon Concentrations

Field use of two different detector types has been demonstrated around an operating uranium mine/mill complex and in approximately 30,000 indoor locations. The results from the measurements around the uranium mine/mill complex indicate that the average monthly radon concentrations change by significant amounts (a factor of 5 to 10) and that the average concentration decreases away from the mine site as might be expected. The highest radon concentrations near the mill were below the concentrations measured in many homes. The results from indoor measurements indicate that the radon concentrations in homes may be significantly influenced by their location with regard to the amount of uranium in the soil and rocks beneath and around the house, as well as the uranium content of the construction materials and the construction of "tightness". In a large percentage of homes already monitored, the radon concentrations exceed provisional indoor standards and a few have radon concentrations that are unacceptable in uranium mines. This work also demonstrates that the radon concentrations in significant numbers of homes exceed 20 pCi/l (0.1 WL). This concentration approaches the maximum permitted exposure in uranium mines. Should current attempts to reduce the 4 WLM mine limit to 0.7 WLM be successful, up to 50 percent of homes monitored in the current work would exceed the mine standard. A significantly larger amount of indoor radon data will be required in the U.S. to establish a reasonable data base and to determine if radon is a major public health problem. The magnitude of the potential problem will become greater as there is greater emphasis on energy conservation in houses and they are sealed for greater energy efficiency.

ACKNOWLEDGEMENTS

This paper could not have been written without the kind cooperation of the following organizations, under whose sponsorship the work was done: U.S. Environmental Protection Administration, Las Vegas, Nevada; Statens Provningsanstalt, Boras, Sweden; General Electric Company, Fairfield, Connecticut; and the Pennsylvania Power and Light Company, Allentown, Pennsylvania.

References

1. Alter, H.W. and R.L. Fleischer 1981, "Passive Integrating Monitor for Environmental Monitoring," Health Physics Vol. 40, pp. 693-702.

2. Alter, H.W. and P.B. Price, 1972, "Radon Detection Using Track Registration Material," U.S. Patent 3,665,194.

3. Gingrich, J.E. and J.C. Fisher, 1976, "Exploration for Uranium Utilizing the Track Etch Technique," 25th International Geological Congress, Sydney, Australia.

4. King, C.Y., 1978, "Radon Emanation on the San Andreas Fault", Nature, Vol. 271, p. 516.

5. Alter,. H.W., 1981, "Passive Integrating Radon Monitor for Environmental Monitoring," 2nd Special Symposium on the Natural Radiation Environment, Bombay, India.

6. Gingrich, J.E., 1980, "Field Measurements with the Passive Integrating Track Etch System," 3rd Symposium on Uranium Mill Tailings Management, Colorado State University, Fort Collins, Colorado. p. 505.

7. Ward, W.J., 1977, "A Convenient Method for Reducing the Radon-220 Background in Uranium Exploration," U.S. Patent 4,064,436.

8. Ward, W.J., R.L. Fleischer and A. Mogro-Campero, 1977, "Barrier Technique for Separate Measurements of Radon Isotopes," Rev.Sci. Instrum. V. 48, p. 1440.

9. McGregor, R.G. et.al., 1980, "Background Concentrations of Radon and Radon Daughters in Canadian Homes," Health Physics, V. 39, p. 285-289.

10. Stranden, E., et.al., 1979, "A Study on Radon in Dwellings," Health Physics, V. 36, p. 413-421.

11. Prichard, H.M., Gesell, T.F., Hess, C.T., Weiffenbach, C. and Nyberg, P., (1981) "Integrating Radon Detector Data from Dwellings in Maine and Texas," EPA International Meeting on Radon and Radon Progeny Measurement. Montgomery, Alabama, August 1981.

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ASSESSMENT OF RADON MEASUREMENT TECHNIQUES USED FOR URANIUM EXPLORATION

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ABSTRACT

Bendix Field Engineering Corporation (BFEC), the prime contractor for the Department of Energy's (DOE) National Uranium Resource Evaluation (NURE) program, has performed studies to characterize and evaluate radon measurement techniques used for uranium exploration. These studies have been performed in the laboratory and in the field at known uranium deposits. The BFEC radon calibration unit has been the principal instrument in this work because it provides a controlled radon atmosphere with which the calibration factor, detection limit, sensitivity, and reproducibility can be determined for various radon measurement devices. The reliability of the measurement techniques is also ascertained by field studies. Radon measurement devices studied have been field-portable zinc sulfide systems, alpha-track detectors, thermoluminescence detectors, diffused-junction detectors, and activated charcoal canisters.

The calibration unit used for these studies includes a 700-liter chamber through which radon in continuously pumped. The radon concentration within the chamber can be varied from 5 to 500 picoCuries per liter (pCi/l) and is continuously monitored. Relative humidity in the chamber can be maintained at a constant level with a microprocessor controller. A radon measurement device can either be placed directly in the chamber or a gas sample can be drawn from the chamber into the device.

Activated charcoal canisters have the lowest detection limit (0.3 pCi/l) and the greatest sensitivity (0.1 pCi/l). One type of activated charcoal canister has a coefficient of variation of 5 percent for replicate samples from the calibration unit. The coefficient of variation for the thermoluminescence detectors is 42 percent. The coefficient of variation measured for all other devices ranges from 11 to 20 percent. The thermoluminescence detectors have the highest detection limit at greater than 30 pCi/l. The portable zinc sulfide detectors have the lowest sensitivity at 2 PCi/l. Radon measurements in the field show more variability than those obtained with the controlled atmosphere in the calibration unit. Based on these studies, one type of activated charcoal canister is superior to the other devices for radon measurements used in uranium exploration.

INTRODUCTION

As part of the NURE program, BFEC has conducted research and development of new and improved uranium exploration techniques and systems. One such uranium exploration technique is the use of near-surface radon measurements to locate subsurface uranium mineralization. As part of the assessment program to determine the usefulness of radon for uranium exploration, a number of radon measurement techniques were used. In order to compare the performance of the various techniques, it was necessary to calibrate (or normalize) them to a common base and to determine their detection characteristics. This was done by studies performed in the laboratory and in the field at known uranium deposits.

It should be emphasized that the techniques discussed in this report were used according to exploration procedures. In this mode one is concerned with relative values and the distinction of anomalous radon measurements from a geochemical background value for an area. Thus, accuracy and precision have been balanced against time, ease of operation, field reliability, and cost in formulating a measurement technique. For many of these techniques one can easily modify the measurement time, detector area or volume, or handling procedure to improve their performance for making environmental measurements. Hence, while the results presented in this report are a fair assessment of the characteristics of a radon technique used in a uranium exploration mode, one needs to be careful in predicting the performance of a technique modified for environmental monitoring.

The BFEC radon calibration unit has been the principal instrument in the assessment work. The unit includes a 700-liter chamber through which air containing radon is continuously pumped. A microprocessor controller is used to maintain the radon concentration and relative humidity within the chamber at constant levels. The radon concentration within the chamber can be varied from 5 to 500 picoCuries per liter (pCi/l) and the relative humidity from 5 to 95 percent. The calibration unit thus provides a controlled radon atmosphere with which the calibration factor, detection limit, sensitivity, and reproducibility can be determined for a radon measurement device.

Field studies were also performed to assess the reliability of a radon measurement technique for a natural situation. Radon measurements were taken at a site of known subsurface uranium mineralization in the Red Desert area of south-central Wyoming. At a sampling location, radon measurements were made for duplicate samples taken 1-meter apart. The simple layered geology of the site and sandy soil cover made this a reasonable approach for determining the influence of the natural environment on a radon measurement technique.

The radon measurement devices studied were field-portable zinc sulfide systems, alpha-track detectors, thermoluminescence detectors, diffused-junction detectors, and activated charcoal canisters. The field-portable, zinc sulfide systems were of the RD-200 type produced by EDA Electronics, Ltd., of Ottawa, Canada, and of the RE-350 type produced by TSA, Inc., of Boulder, Colorado. The alpha-track detectors were Track Etch® cups produced by Terradex Corporation of Walnut Creek, California. The thermoluminescence detectors were Alpha-2™ cups manufactured by Westinghouse

Electric Corporation of Pittsburgh, Pennsylvania. The silicon diffused-junction detectors were alphaMETERS produced by alphaNUCLEAR Company of Mississauga, Ontario, Canada. Two types of activated charcoal canisters were studied. One was a modified U.S. Army M11 gas canister obtained from Union Carbide at Oak Ridge, Tennessee, and the other was ROAC cups produced by Inter-Science Research and Development Services (Pty) Ltd., of Johannesburg, South Africa.

RADON CALIBRATION UNIT STUDIES

The BFEC radon calibration unit is shown in Figure 1. It has been the principal instrument in assessing the performance of radon measurement devices. Radon is generated in four ion-exchange filters by the radioactive decay of radium deposited on the filters. The radon from one or more sources is swept along in airflow of 0.1 to 0.2 liters per minute and is continuously delivered into the radon chamber. A larger airflow of 5 to 20 liters per minute of filtered air containing variable humidity is introduced into the chamber simultaneously. The air in the chamber is exhausted through a vent outlet on the top of the building in which the calibration unit is located. Thus, the calibration unit is a dynamic system and radon-daughter buildup is minimized.

The radon concentration and relative humidity in the chamber can be maintained at a constant level with a microprocessor controller. The radon activity in the chamber is monitored with a continuous flow-through zinc sulfide scintillation detector. Air from the chamber is pumped at 1.5 liters per minute through the detector and returned to the chamber. The output of the detector is used by the microprocessor controller to adjust flowrates so that the radon concentration in the chamber only varies by 2 percent. The radon concentration within the chamber can be varied from 5 to 500 picoCuries per liter (pCi/l). The relative humidity in the chamber is monitored with a Hygrocon II relative humidity unit. Using the output of this device, the microprocessor controller has the dilution air for the chamber going through either a high humidity or a heatless drying cycle prior to entering the chamber. Under microprocessor control, the relative humidity in the chamber varies by less than 1 percent. The temperature and pressure within the chamber are also monitored, but are not controlled.

The calibration chamber is a large fiberglass glove box with an internal volume of 700 liters. It is equipped with an interchange box that allows the insertion of radon devices to be studied into the interior without significantly disturbing the steady-state condition of the chamber. Figure 2 illustrates the large number of devices that can be placed directly in the chamber at a single time. The air in the chamber is well-mixed and the radon is uniformly distributed even when a significant volume of the chamber is occupied by radon devices. It is also possible to draw a gas sample from the chamber as shown in Figure 3.

The calibration of the unit is currently based on calibration factors obtained at the U.S Department of Energy's Environmental Measurements Laboratory (EML). These factors were obtained by calibrating a group of zinc sulfide scintillation flasks and our counting system at EML. Figures 4 and 5 show this calibration procedure.

Figure 6 lists the characteristics of the radon calibration unit used to assess radon measurement techniques used for uranium exploration. This list summarizes the previous discussion of the unit. In using the unit for the assessment studies, the unit was always operated with the relative humidity maintained at 30 percent. For most studies the radon concentration was maintained at approximately 100 pCi/l. One aspect of the techniques that varies considerably is exposure or collection time. While one could treat exposure time as a separate variable, the author has chosen to define it as fixed at the manufacturer's recommended exposure time and as an integral part of a technique.

The calibration factor for each device was determined with the calibration unit. This was done with each device operated in the manner used for uranium exploration. These calibration factors provide a common basis for determining the detection limit, sensitivity, and reproducibility of a radon measurement device. The detection limit of a device is the radon concentration equal to the inherent background level of the device. This was determined by measuring the signal present in devices that had not been exposed to any radon. The sensitivity of a device is the minimum variation in radon concentrations that is detectable by the device. This was determined by the calibration factor for a device and a device's minimum unit of measure. The reproducibility was determined for replicate radon measurements and is indicated by the coefficient of variation which is the standard deviation of the replicate measurements divided by the mean of the measurements and multiplied by 100 percent.

FIELD STUDIES

In order to ascertain the reliability of a radon measurement technique in the natural environment, radon measurements were made at sites of known subsurface uranium mineralization. The studies for only one site, the Red Desert area of south-central Wyoming, are presented in this paper. The number of techniques used at this site was more comprehensive than the other sites and the geologic setting is fairly simple. Descriptions of the site can be found in Bramlett et al., 1981 and Sherborne et al., 1979. The site is situated on a gently rolling plain that generally dips to the south. The soil is essentially sandy and contains primarily quartz and feldspar grains derived from the underlying sandstone of the Battle Spring Formation. The site is underlain by a sequence of interfingered sandstone, shale, and coal layers. The known deposit is a roll-front deposit that lies within an arkosic sand at a depth of 140 meters.

The reliability of a radon measurement technique was determined by making duplicate radon measurements 1 meter apart at a sampling location. Thirty-four locations on a grid with spacings of 300 meters were designated at the site. The radon measurements were made 0.7 meters below the surface. This is a typical sampling depth for radon measurements performed for uranium exploration. Soil gas at this depth was obtained with a 6-millimeter-diameter probe for the portable zinc sulfide systems. The other radon measurement devices were placed in 0.15-meter-diameter holes at a depth of 0.7 meters.

RADON MEASUREMENT TECHNIQUES

The radon measurement techniques studied used field-portable zinc sulfide systems, alpha-track detectors, thermoluminescence detectors, diffused-junction detectors, and activated charcoal canisters. Descriptions of these techniques are found in Pacer and Czarnecki, 1980. The field-portable zinc sulfide systems are shown in Figure 7. They are the RD-200 produced by EDA and the RE-350 produced by TSA. The RD-200 uses a bulb pump to acquire a gas sample. The alpha activity in the zinc sulfide chamber is counted for three consecutive 1 minute intervals. These counts are used to compute the radon count rate according to a formula developed by Morse (1976) which accounts for the presence of thoron and the ingrowth of radon daughters. The RE-350 is similar to the RD-200, but it has a microprocessor controller which controls a diaphragm pump for consistent sample collection and the subsequent alpha activity counting. The microprocessor can then subtract background, correct for thoron decay and radon daughter ingrowth, and use its calibration factor to display the radon concentration of the gas sample directly in pCi/l.

The alpha-track detectors and thermoluminescence detectors are shown in Figure 8. The alpha-track detectors are Track Etch ® cups produced by Terradex, that employed a polycarbonate track material. The thermoluminescence detectors are Alpha-2 ™ cups manufactured by Westinghouse. Both types of cups are typically exposed for 30 days in a uranium exploration mode. They are then returned to their respective manufacturers for processing. The radon exposure for the Track Etch ® cups is reported in tracks per square millimeter for 30 days (T/mm^2-30d). The radon exposure for the Alpha-2 ™ cups is reported as net counts for 30 days.

A silicon diffused-junction detector is shown in Figure 9. The device is an alphaMETER produced by alphaNUCLEAR. This device detects and integrates alpha particles which impinge the surface of the detector. In a uranium exploration mode, these devices are typically exposed for 1 to 3 days at a location.

Two types of activated charcoal canisters are shown in Figure 10. They are the M11 canisters and the ROAC cups. We modified the procedure for using the M11 canisters described by Countess, 1976, for uranium exploration. The canisters are exposed for 2 days, held for at least 60 hours for thoron-daughter decay, and then gross-gamma counted for 15 minutes. This procedure is also used for the ROAC cups, though the manufacturer recommends a 1-minute gross-gamma measurement of the ROAC cartridge immediately on retrieval.

Figure 11 summarizes the radon measurement techniques.

RESULTS

The results of the studies performed with the radon calibration unit are shown in Figure 12. In terms of detection limits, all the devices except the Alpha-2 ™ detectors have reasonably low detection limits. The charcoal canisters have the lowest detection limit. The thermoluminescence detector material used in the Alpha-2 ™ cups is sensitive to radiation other than alpha particles and its background is continually increasing. Earlier studies by the author with Track Etch ® cups indicated that they have a detection limit of 100 pCi/l, but improvements by the manufacturer have reduced this to 1.7 pCi/l.

All the measurement techniques are very sensitive to measuring radon. The activated charcoal canisters have the greatest sensitivity and the portable zinc sulfide systems have the lowest sensitivity.

For replicate measurements in the controlled environment of the calibration unit, most devices had a coefficient of variation from 11 to 20 percent. The exceptions were the ROAC™ cups with a coefficient of variation of only 5 percent and the Alpha-2™ cups which were at 42 percent. The results of the replicate measurements made at the field site are shown in Figure 13. Unfortunately, the ROAC cups and the alphaMETERS were not used at the field site. So, there is no certainty that the ROAC cups would perform as well in this environment. The two portable zinc sulfide systems responded differently to the field environment. While the TSA RE-350 emanometer was unaffected by the field environment, the coefficient of variation for the EDA RD-200 increased from 11 percent in a laboratory environment to 31 percent in a field environment. This increase in variability probably reflects variability in the operation of a device of this complexity when the device is being used in the field. The RE-350 emanometer did not demonstrate this variability since it is microprocessor controlled. The Track Etch® cups and M11 charcoal canisters exhibit some decrease in reliability in going to a field environment. The Alpha-2™ cups show improved reliability for the field environment. This probably is due to the higher radon concentrations (mean radon concentration of about 2000 pCi/l) encountered in the soil gas of the site compared to the BFEC radon calibration unit.

CONCLUSIONS

The detection limit, sensitivity, and reproducibility were determined for various radon measurement devices by studies performed in the laboratory with the BFEC radon calibration unit and in the field. Based on these studies, one type of activated charcoal canister, the ROAC cup, is superior to the other devices for radon measurement used in uranium exploration.

REFERENCES

Bramlett, L., Pacer, J.C. and Moll, S., 1981, Geochemical exploration for uranium in the Red Desert, Wyoming: U.S. Department of Energy Open-File Report GJBX-125(81), 40 p.

Countess, R.J., 1976, 222Rn flux measurement with a charcoal canister: Health Phys., v. 31, p. 456-457.

Morse, R.H., 1976, Radon counters in uranium exploration: Exploration for uranium ore deposits, Vienna, International Atomic Energy Agency, p. 229-239.

Pacer, J.C. and Czarnecki, R.F., 1980, Principles and characteristics of surface radon and helium techniques used in uranium exploration: U.S. Department of Energy Open-File Report GJBX-177(80), 64 p.

Sherborne, J.E., Pavlak, S.J., Peterson, C.H., and Buckovic, W.A., 1979, Uranium deposits of the Sweetwater Mine area, Great Divide Basin, Wyoming: Regional AIME meeting, Casper, Wyoming, September, 1979.

FIGURES

1. BFEC radon calibration unit: the rack (left) contains recorder for various sensors, microprocessor controller, flowmeters, and radium sources and the glove box (right) is the radon chamber.

2. View of inside of radon chamber containing many Track Etch cups and an operator holding an alphaMETER.

3. RE-350 radon emanometer being tested with the BFEC radon calibration unit.

4. Zinc sulfide cells being filled with the EML radon chamber.

5. The zinc sulfide cells being counted at EML.

6. List of radon calibration unit specifications.

7. Portable zinc sulfide systems: the RE-350 (left) is produced by TSA, Inc. and the RD-200 (right) is produced by EDA Electronics.

8. Alpha track detectors, Track Etch cups (bottom row), are produced by Terradex Corporation and thermoluminescence detectors, Alpha-2 cups (top row), are produced by Westinghouse Electric Corporation.

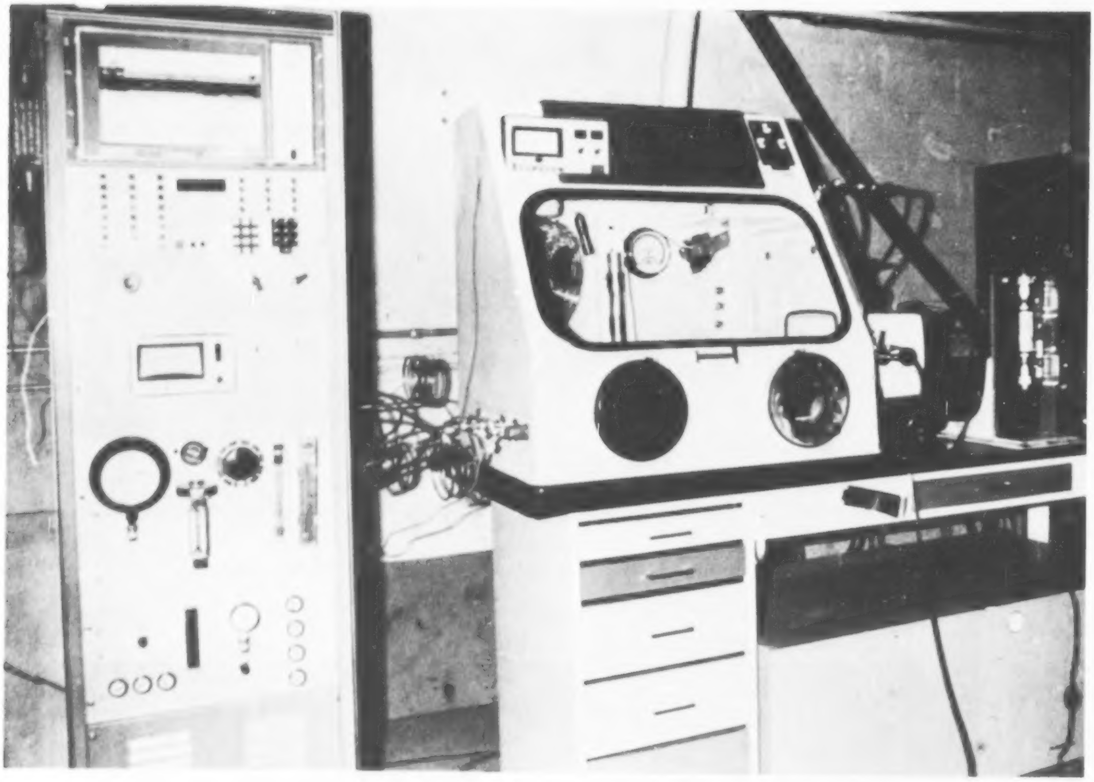
9. A silicon diffused junction device, the alphaMETER (left) is produced by alphaNUCLEAR Company and a recorder (right) for ten simultaneously used devices.

10. Activated charcoal devices: the ROAC cup (left) and the ROAC cartridge (center) are produced by Inter-Scierce Research and Development and the M11 canister (right) is obtained from Union Carbide.

11. List of radon measurement techniques used for uranium exploration.

12. Characteristics of radon detectors determined with the radon calibration unit.

13. Reliability of radon measurement techniques determined by field studies.



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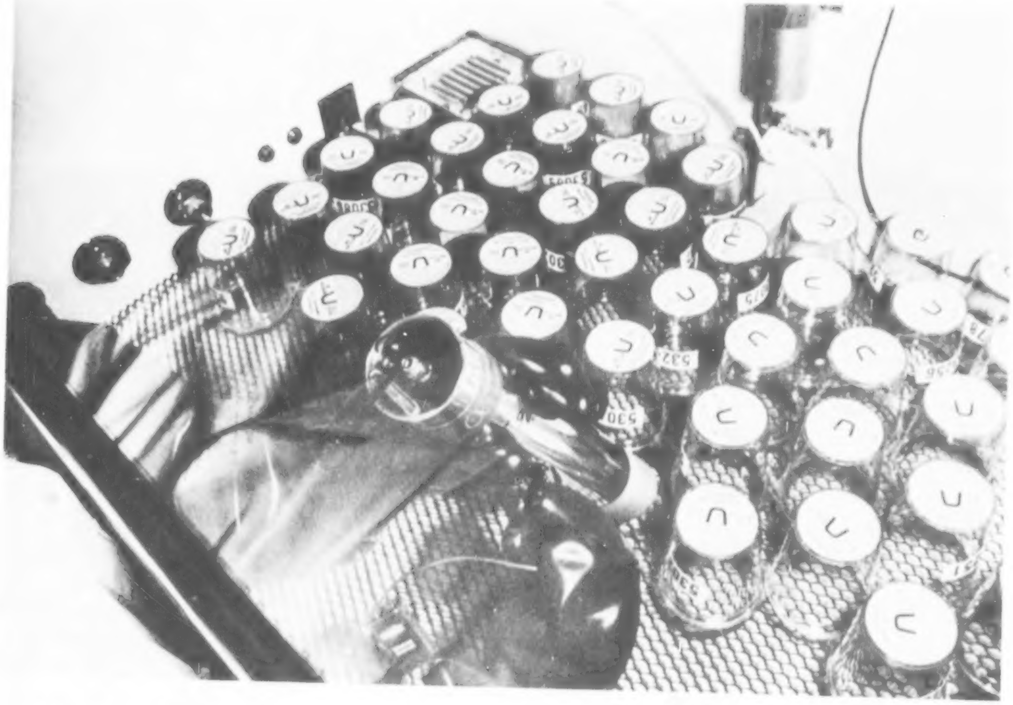

Figure 2. View of inside of radon chamber containing many Track Etch cups and an operator holding an alphaMETER.

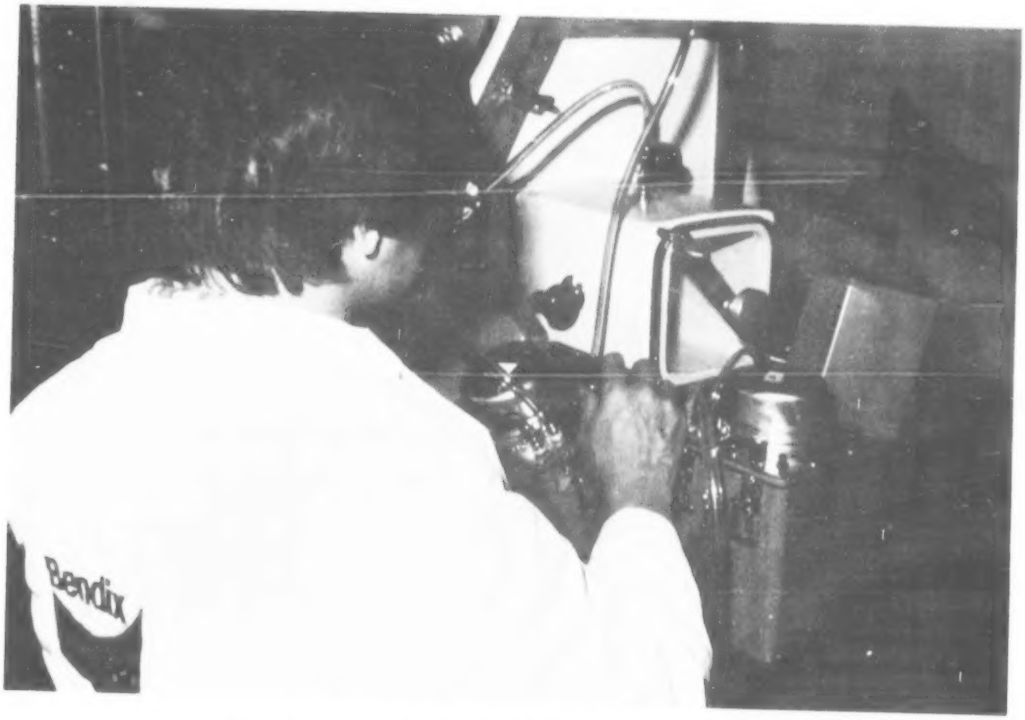

Figure 3. RE-350 radon emanometer being tested with the BFEC radon calibration unit.

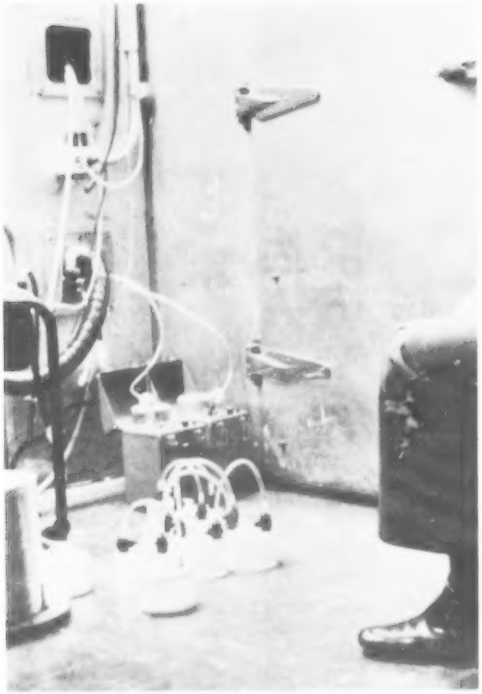

Figure 4. Zinc sulfide cells being filled with the EML radon chamber.

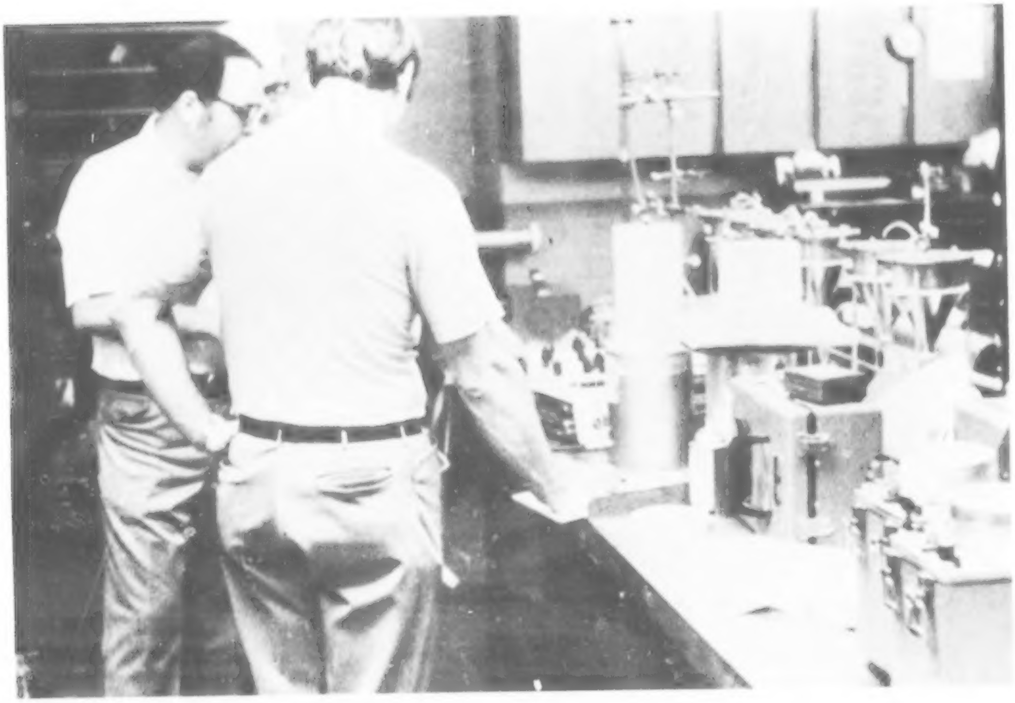

Figure 5. The zinc sulfide cells being counted at EML.

Radon Calibration Unit Specifications

1. Dynamic System
2. Chamber Volume 700 Liters
3. Microprocessor Controlled Humidity: 5-95%
4. Radon Concentration Range: 5-500 pCi/liter
5. Temporal Radon Variation:
    a. Hourly: ±2.6% at 60p Ci/liter mean
    b. Daily: ±2.2% at 60p Ci/liter mean

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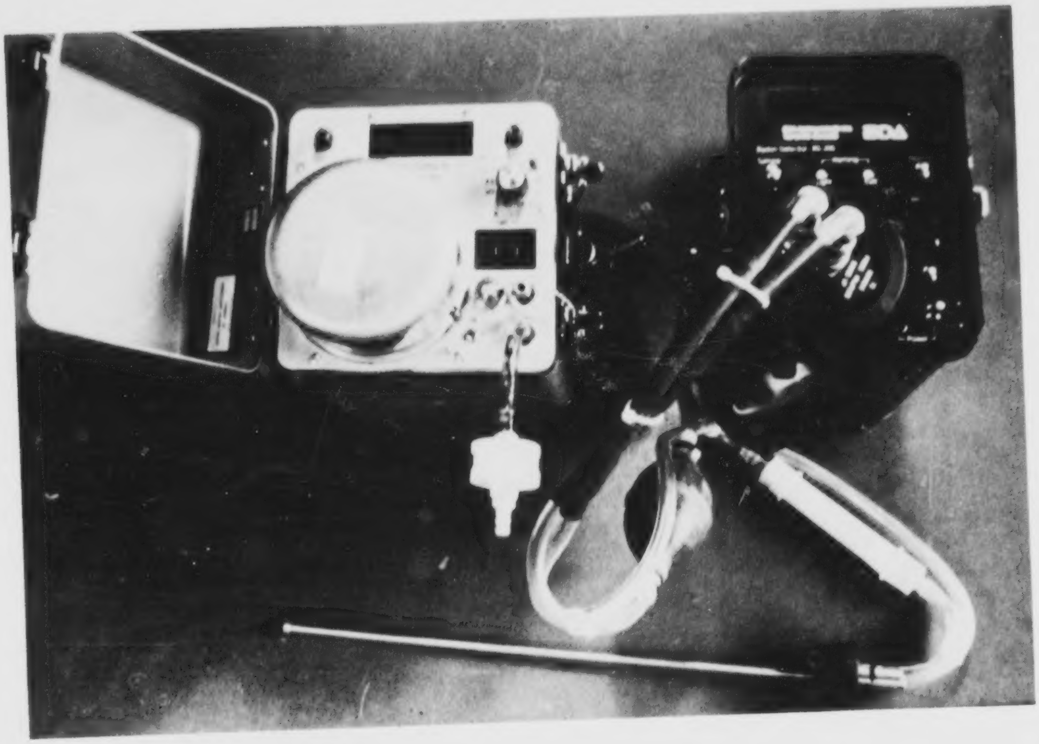
Figure 6. List of radon calibration unit specifications.



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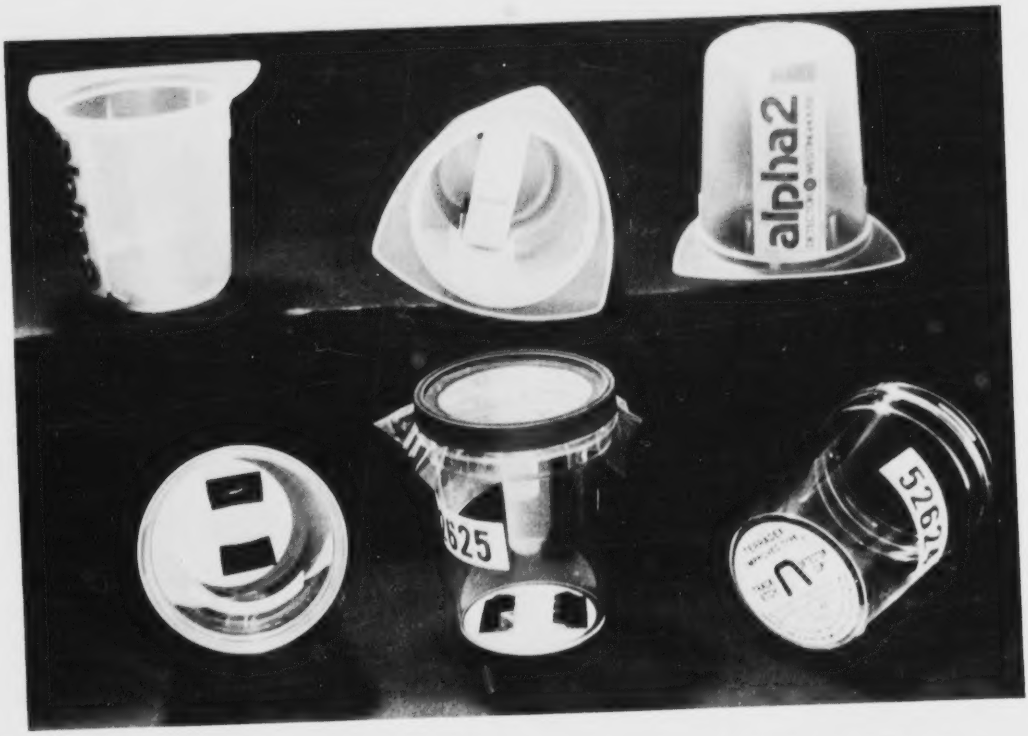

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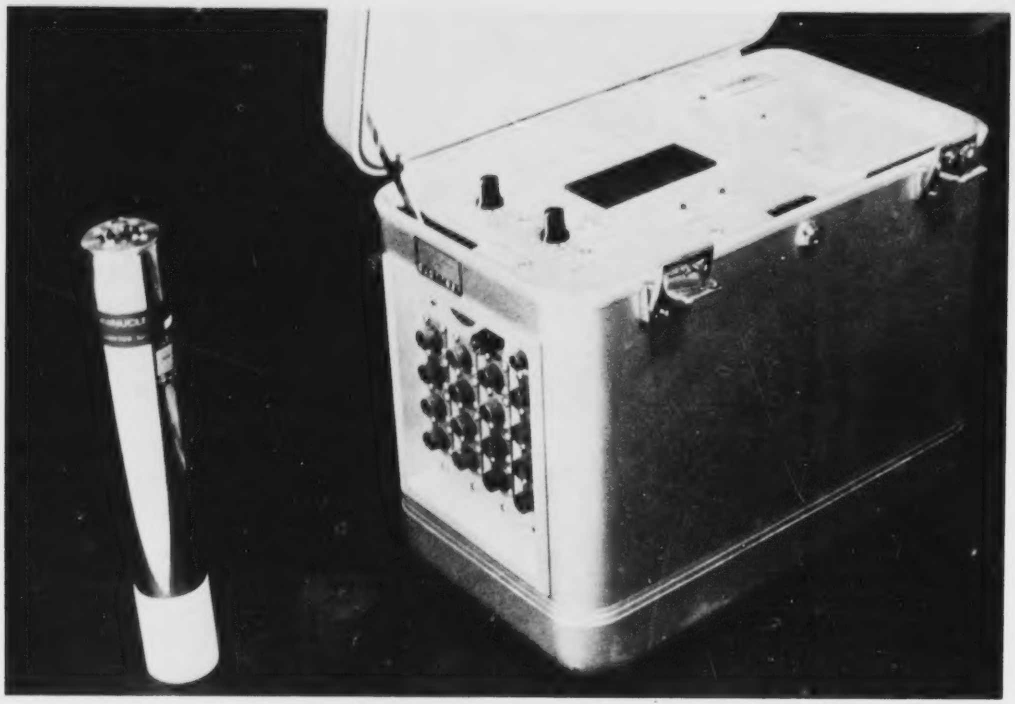

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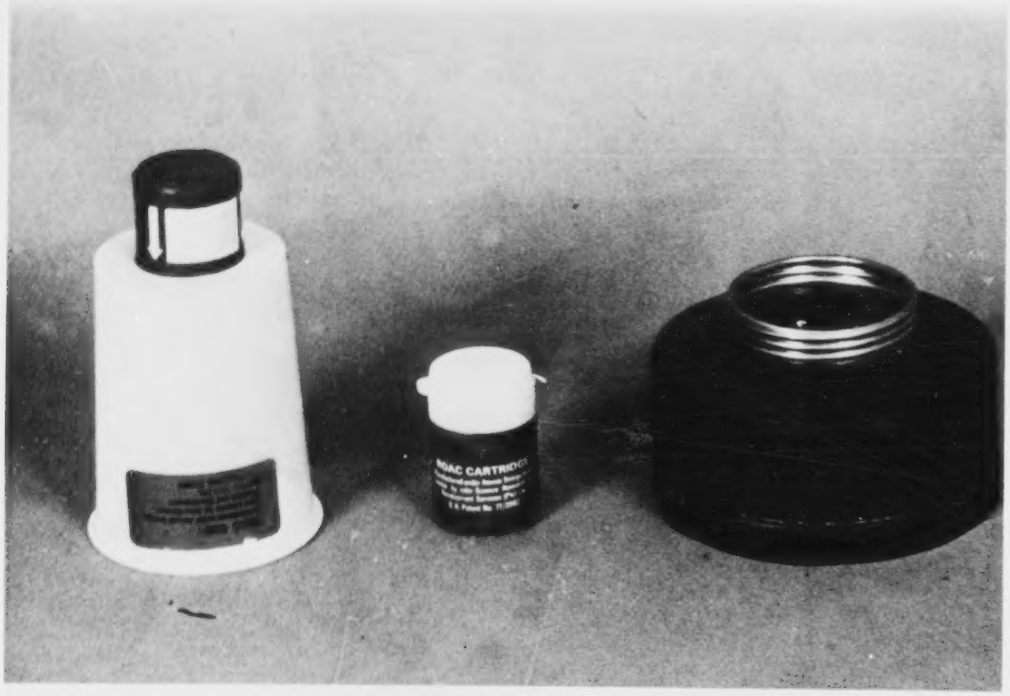

Figure 10. Activated charcoal devices: the ROAC cup (left) and the ROAC cartridge (center) are produced by Inter-Science Research and Development and the M11 canister (right) is obtained from Union Carbide.

Radon Measurement Techniques Used for Uranium Exploration

Technique	Device	Sample Collection or Exposure Time
Portable Zinc Sulfide System	EDA RD-200	10 minutes
	TSA RE-350	15 minutes
Alpha Track Detector	Track Etch Cup	30 days
Thermoluminescence Detector	Alpha - 2 Cup	30 days
Diffused Junction Detector	Alpha METER	2 days
Activated Charcoal Canister	M11	2 days
	ROAC Cup	2 days

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Figure 11. List of radon measurement techniques used for uranium exploration.

Characteristics of Radon Detectors Determined With the Radon Calibration Unit

Detector	Detection Limit (pCi/l)	Sensitivity (pCi/l)	Coefficient of Variation (%)
EDA RD-200	12.0	2.1	11
TSA RE-350	13.0	2.2	13
Track Etch	1.7	0.3	16
Alpha - 2	30.4 & 2.4/month	0.3	42
Alpha METER	1.3	0.5	20
M11 Charcoal Canister	1.0	0.1	12
ROAC	0.3	0.1	5

1048-8/81

Figure 12. Characteristics of radon detectors determined with the radon calibration unit.

Reliability of Radon Measurement Techniques Determined by Field Studies

Detector	Coefficient of Variation (%)
EDA RD-200	31
TSA RE-350	13
Track Etch	22
Alpha - 2	15
M11 Charcoal Canister	23

1045-8/81

Figure 13. Reliability of radon measurement techniques determined by field studies.

A REVIEW OF SOME EXACT METHODS FOR ^{222}RN-DAUGHTER MEASUREMENT*

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ABSTRACT

Some exact methods for the measurement of the short-lived ^{222}Rn progeny are discussed. All methods use three separate measurements of alpha activity or a combination of alpha activity and total beta activity. The latter methods permit a rapid determination of individual Rn-daughter concentrations with several Instant Working Level Meters (IWLM) and Environmental Working Level Monitors. The former methods, based only on the measurement of alpha activity, were originally used for the calibration of these devices but can, of course, be used separately. They differ from the usual methods in their counting philosophy. Alpha counts accumulated during the first counting interval are recorded, and counts from subsequent intervals are added. The resulting larger total counts yield greater statistical precision than similar methods that reset the counter at the end of each counting interval. A brief discussion of the probabilistic treatment of Rn-daughter build-up and decay on a filter paper and some comments on counting statistics conclude the paper.

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*This paper is based on work performed under Contract No. DE-AC05-760R00033 between the Department of Energy, Office of Health and Environmental Research, and Oak Ridge Associated Universities.

TRACK ETCH CALIBRATION:
DESIGN AND RESULTS OF A PILOT STUDY

Susan M. Hinkins

INTRODUCTION

The Office of Radiation Programs will be conducting a field study of radon and radon progeny measurement techniques. One of the measurement devices used will be the Track Etch Device * (TED). Prior to their deployment in the field we will run a calibration study of the TED's in a radon chamber. This paper briefly describes a pilot study done in anticipation of this calibration study.

The purpose of the pilot study is to gather information which will help us anticipate and avoid problems which might otherwise occur in the main study. A successful pilot study will identify problems in data quality control which can then be corrected before the main study begins. This study identified 3 data problems:

- the data contain at least one outlier.
- the number of tracks at the highest exposure is substantially lower than the number of tracks at the next highest exposure.
- there appears to be a "family resemblance" in data from devices exposed and read at the same time.

THE STUDY DESIGN

The independent variable is the total exposure to radon, which is measured as the average radon concentration (p Ci/l) times the exposure time (days). Because the radon chamber was being held at an approximately constant radon level, we could vary the exposure only by varying the exposure time. Therefore total exposure is confounded with exposure time; in the calibration study we would avoid this by varying both radon level and exposure time. The average radon concentration in the chamber was calculated from hourly measurements from flow-through radon gas monitors. Scintillation cell measurements were taken several times each day, also.

The response measure on the TED's is the total number of tracks in 100 microscope fields (approximately 5.75 mm^2). For a given exposure, X, we expect the total counts, T, to be distributed as a Poisson variable with mean proportional to exposure. Therefore we expect the relationship to be T = bX, a straight line through the origin. The Poisson distribution has the property that the variance is equal to the mean.

The chamber was run for 38 days and six exposures were planned: 3, 4, 7, 14, 21 and 35-day exposures. We later included 38-day measurements because they were available.

Unfortunately, we failed to specify the placement of the devices in the chamber, so exposure is confounded with location in the chamber. We will not make the same mistake in the calibration study.

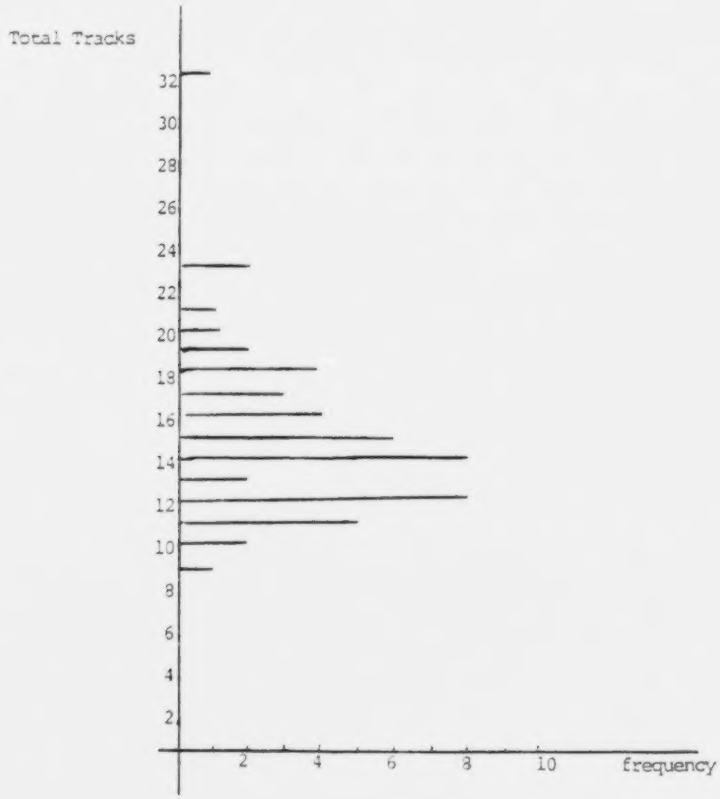
All TED's were filter cups from batch 8.

AN OUTLIER

The largest value in the 3-day data is an outlier. Assuming all 3-day values come from the same Poisson distribution, the probability of getting such a large value is less than .0001.

We found no reason to believe that this extreme value was due to mislabeling or other clerical errors. The plot of the 3-day data is given in Figure 1.

Figure 1



WHAT'S WRONG WITH THE 35 AND 38-DAY DATA?

The data at exposures less than 35 days can be fit very well by a straight line through the origin. However, the number of tracks at the 38-day exposure is significantly smaller than the number of tracks at the 35-day exposure.

Again, we could find no evidence of clerical error. After investigating several possible explanations we were left with the following two unresolved possibilities.

i) While we do not believe that the location of the devices in the chamber could cause such an effect, we cannot discard this possibility because the exposure groups were confounded with location. A study currently underway may resolve this question.

ii) Since the 35 and 38-day TED's were sent in at different times, they were presumably etched at different times. Possibly the etching process can cause this much variation.

THE "FAMILY RESEMBLANCE"

What originally interested us in the possibility of an etching or reading effect was that the 35-day values, which were higher than expected, were read at the same time as the highest 7-day readings and the highest 4-day readings. For a given exposure, 3, 4, or 7-day, we looked at blocks of TED's, where a block is defined as the set of TED's which were exposed together, mailed together, and presumably etched and read together. When we examined the data by blocks, we found that the sample variance within blocks is often much smaller than expected for a Poisson model. Also one-way analyses of variance show significant differences between blocks.

As an example, the summary of the 7-day data is given in Figure 2. Recall that under the assumption of a Poisson distribution, the variance should equal the mean.

A smaller variance than expected indicates an error in the assumptions, or an unexplained and uncontrolled source of correlation and variability. This source could be a reader effect, smoothing the data toward the mean.

Figure 2

7-day Series

Block	Sample Mean	Sample Variance
1	34.0	6.5
2	30.2	4.7
3	29.6	10.3
4	29.6	5.3
5	36.4	19.8
All data	31.96	15.71

1 way Analysis of Variance

Source	d.f.	S.S.	MS
Between	4	190.6	47.6
Within	20	186.4	9.3
C. Total	24	377.0	

SUMMARY

The pilot study identified three data problems.

1) The 3-day data contain one extreme value. While this is not cause for concern, it is important to assess the likelihood of outliers before deploying the devices.

2) Although at the five lower exposures (21 days or less) the number of tracks is proportional to exposure, the number of tracks at the highest exposure (38-days) is substantially lower than the number of tracks at the next highest exposure (35 days). Could this be due to an etching or other procedural effect?

3) Measurements from TED's exposed and read at the same time vary substantially less than would be expected under a Poisson model. Measurements from TED's with similar exposures but read at different times vary more than would be expected, but this may be caused in part by the small variance within blocks. This "family resemblance" may be due to the etching process or to a reader error, a smoothing effect.

MEASURING RADON SOURCE MAGNITUDE IN RESIDENTIAL BUILDINGS*

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August 1981

ABSTRACT

We describe the procedures we use in residences for rapid "grab-sample" and time-dependent measurements of the air-exchange rate and radon concentration; the radon source magnitude is calculated from the results of simultaneous measurements of these parameters. Grab-sample measurements in three survey groups comprising 101 U.S. houses showed the radon source magnitude to vary approximately log-normally with a geometric mean of 0.37 and a range of 0.01 to 6.0 pCi l^{-1} h^{-1}. Successive measurements in six houses in the northeastern United States showed considerable variability in source magnitude within a given house; in two of these houses the source magnitude showed a strong correlation with the air-exchange rate, suggesting that soil gas influx can be an important transport process for indoor radon.

Keywords: radon, houses, air-exchange rate, pollutant sources.

INTRODUCTION

Characterizing radon sources and the processes by which radon is transported into buildings is an important component in developing a comprehensive understanding of radon as a contaminant of indoor air. For sources such as building materials, domestic water, and natural gas, characterization is relatively straightforward; however, these sources do not appear to contribute to indoor radon concentrations in amounts sufficient to account for the levels observed in a large number of houses surveyed in the United States and Canada. Rather, evidence is mounting that local rock and soil (referred to simply as "soil" throughout this paper) are the dominant sources of radon in these houses. The characterization of soil as a source of indoor radon is difficult, because, for example, the permeability and radon emanating power of soil vary with changes in moisture content, radon can migrate over long distances in soil; and radon can enter a house by either bulk flow or diffusion.

A potentially important approach in investigating the origin of radon indoors is to calculate the effective radon source magnitude by simultaneously measuring the radon concentration and the air-exchange rate in a house. It is the initial results obtained from such measurements that strongly suggest soil to be the dominant source of radon in a significant proportion of U.S. houses. Presuming this to be the case, we can use measurements of radon source magnitude to ascertain the potential for high radon levels in a geographic area. Furthermore, by measuring radon source magnitude continuously in conjunction with meteorological parameters such as wind speed, indoor-outdoor temperature difference, and barometric pressure, we can gain a better understanding of processes which transport radon from its source material to indoor air.

To date, we have measured radon source magnitudes in about 100 U.S. houses. In most of these homes we used a single simultaneous measurement of radon concentration and air-exchange rate to determine the radon source magnitude at one point in time; however, in several we made successive measurements of the radon source magnitude over periods of time ranging from 3 to 30 days. Our most significant findings are: (1) that the radon source magnitude varies over a wide range from one house to another; (2) that the radon source magnitude in a house can change dramatically even over a period as short as several hours; and (3) that frequently the radon source magnitude in a single house shows a positive correlation with the infiltration rate -- consistent with the hypothesis that soil is important as a source of indoor radon and that bulk flow is important as a transport mechanism, as we shall discuss.

In this paper we describe the instrumentation and techniques used in our studies to calculate the radon source magnitudes in residences. We present the results of successive measurements of source magnitudes taken in six houses in the eastern part of the United States, and use these along with the results of a grab-sample survey of 101 houses variously located in the United States as a basis for discussing some aspects of the origin and transport of radon in U.S. houses.

METHODOLOGY

Theory

The mass-balance equation for indoor radon can be written as

$$\frac{dR_i(t)}{dt} = S_R(t) + \lambda_v(t)R_o(t) - \lambda_{Rn}R_i(t) - \lambda_v(t)R_i(t) \qquad (1)$$

where $R_i(t)$ is the indoor radon concentration, $R_o(t)$ is the outdoor radon concentration, $S_R(t)$ is the indoor radon source magnitude per unit volume, $\lambda_v(t)$ is the air-exchange rate, and λ_{Rn} is the time constant for radioactive decay of radon (0.00756 h^{-1}). Since λ_v almost always greater than 0.1 h^{-1}) is much larger than λ_{Rn}, we ignore the third term on the right hand side of Eq.(1). We define the "effective" indoor radon source magnitude, Q_R, as the sum of the first two terms on the right hand side of Eq.(1) so that for steady-state conditions we have

$$Q^s_R = S^s_R + \lambda^s_v R^s_o = \lambda^s_v R^s_i \ , \tag{2}$$

where the superscript s denotes the steady-state value. In this way a simultaneous measurement of the air-exchange rate and the indoor radon concentration can be used to compute the effective source magnitude, assuming that steady-state conditions prevail. The difference between the effective source magnitude, Q_R, and the value for indoor sources, S_R, is small if the indoor radon concentration is much greater than the outdoor concentration, as is often the case.

By continuously measuring the radon concentration and the air-exchange rate, we can calculate the source magnitude even under changing conditions. Equation (1) is solved to obtain

$$\langle Q_R \rangle_{0,t'} = \frac{R_i(t') - R_i(0)}{t'} + \langle R_i \lambda_v \rangle_{0,t'} , \tag{3}$$

where $\langle\rangle_{0,t'}$ indicates a time-weighted average over the interval 0 to t' of the contents of the brackets. Because we measure radon averaged over finite time intervals, we do not know $R_i(t')$ or $R_i(0)$. We approximate these two quantities as

$$R_i(t') \approx 1/2\ (\langle R_i \rangle_{0,t'} + \langle R_i \rangle_{t',2t'}) \ ,$$

and

$$R_i(0) \approx 1/2\ (\langle R_i \rangle_{-t',0} + \langle R_i \rangle_{0,t'}) \ . \tag{4}$$

We further approximate

$\langle R_i(t)\ \lambda_v(t) \rangle_{0,t'}$ by $\langle R_i(t) \rangle_{0,t'} \langle \lambda_v(t) \rangle_{0,t'}$.
This approximation is accurate as long as there is no correlation between R_i and λ_v, or as long as both of these parameters do not vary significantly during a measurement interval. Assuming a continuous measurement of radon concentration, our estimate of the effective radon source magnitude is

$$\langle Q_R \rangle_{0,t'} = \frac{(\langle R_i \rangle_{t',2t'} - \langle R_i \rangle_{-t',0})}{2t'} + \langle R_i \rangle_{0,t'} \langle \lambda_v \rangle_{0,t'} \ . \tag{5}$$

Grab-Sampling: Technique and Instrumentation

Radon

Grab-sample measurements of radon are made using scintillation cells, either cells fabricated in our laboratory after the design of Lucas (Lu57) or commercially-available cells (EDA, Model RDX 388). The procedure used for taking the sample varies: (1) The sample is taken directly with the scintillation cell which is then either counted in the field or sent back to our laboratory for analysis. (2) The sample is taken in a radon-impermeable sampling bag (Environmental Measurements, Inc., air-sampling bag, made of Tedlar) which is sent back to our laboratory and analyzed by transferring a portion of the sample to a scintillation cell. (3) A metal vessel of known volume (typically 1 liter) is used to collect the sample, and then returned to our laboratory where the radon is extracted by passing the sample through a glass-wool trap cooled to -196° C in a liquid N_2 bath; the extracted radon is then transferred to a scintillation cell. This procedure is described by Lucas (Lu77), and our implementation of it is reported by Ingersoll (In80). The procedure for concentrating radon is important for measurement precision and improvement of our sample throughput rate when measuring radon concentrations below about 1pCi/l.

The scintillation cells are calibrated in batches. A calibration factor is determined for the batch by filling several of the cells with a known amount of radon, derived from a standard-reference-method solution of ^{226}Ra (National Bureau of Standards). The response of individual cells within the batch is checked by filling several cells with a constant, unknown concentration of radon, and comparing their count rates. The responses of individual cells fabricated at Lawrence Berkeley laboratory (LBL) have been found to be within a range of 5%. We occasionally check our calibration factor against those independently determined by other laboratories. (In a recent check we were among six of the nine laboratories participating whose calibration factors agreed within a range of 10% (Ge81).)

Air-Exchange Rate

We measure air-exchange rate by tracer gas decay, most commonly using sulfur hexafluoride (SF_6), but occasionally ethane, as the tracer. The tracer gas is injected into the house and mixed to a uniform concentration by means of portable fans or the furnace fan. The concentration is monitored over time and the resulting data are fit to an exponential decay curve of the form

$$C(t) = C(0) \exp(-\lambda_v t) \ , \qquad (6)$$

where the time constant, λ_v, is the air-exchange rate.

SF_6 concentrations are measured with a commercially-available, non-dispersive infrared (NDIR) analyzer (Foxboro-Wilks, Model Miran 101). The analyzer is calibrated by measuring its response to gases derived from compressed-air tanks containing known concentrations of SF_6.

Continuous Measurement: Techniques and Instruments

Radon

The radon concentration is measured over time with a Continuous Radon Monitor (CRM) developed by Thomas (Th79), and designed and fabricated at LBL. Figure 1 presents a schematic diagram of our most recent version of the CRM, whose unique feature is that the output is provided as an analog rather than a digital signal, simplifying the interface of the CRM to recording devices (strip-chart recorder or a data logger). The output voltage has a range of 0 to 1023 mV, corresponding to 0 to 1023 counts in the counter. After reaching 1023 the counter automatically resets. Converting the count total to an analog voltage introduces an uncertainty on the order of one count, which is insignificant compared to the statistical uncertainty in measuring radioactive decay. We use three-hour integration intervals for analyzing the CRM data.

The CRM is calibrated following the procedure of Busigin et al. (Bu79); the radon concentration is calculated as follows:

$$\langle R_i \rangle_{t',t'+3h} = 2.19 \langle n \rangle_{t',t'+3h} - 0.13 \langle R_i \rangle_{t'-3h,t'} , \tag{7}$$

where n is the net count rate in min^{-1}. Aside from calibration uncertainties, the relative standard deviation in the measurement of a steady-state radon concentration is typically 6% at 5 pCi/l, 21% at 1 pCi/l, and 48% at 0.4 pCi/l (Na81).

Air-Exchange Rate

We have developed an automated system, the Aardvark, for continuously measuring the air-exchange rate and the radon concentration in an occupied residence. It is described in detail in another report (Na81). Briefly, the Aardvark uses the tracer gas decay technique with SF_6 to measure the air-exchange rate. The operation of the system is controlled by a microcomputer system, which also performs preliminary data analysis. The data are recorded on magnetic tape and printed on a terminal.

The mechanical system of the Aardvark is presented in Figure 2. As illustrated, a blower is used to draw and return air at a total rate of 80 l/m through as many as four sampling lines and up to four delivery lines. A bypass loop provides 20 l/m to the SF_6 analyzer and 2 l/m to a CRM which is interfaced to the system. The analyzer is automatically calibrated at user-specified intervals by measuring its response to three concentrations of SF_6 drawn from compressed-air tanks.

To ensure good distribution and thorough mixing of the tracer gas in a house with a forced-air furnace, we use only one delivery line, installing its end in the return-air duct of the furnace system. A relay is used to bypass the thermostat and turn on the furnace fan during injection of the tracer gas. In a house that does not have air-distribution ductwork, mixing is accomplished by using three or four delivery lines, attaching the end of each to a portable fan which is turned on during injection. In either case, three or four sampling lines are used to reduce the dependence of the air-exchange rate measurement on the location of the sampling point(s).

Air-exchange rates are measured by the Aardvark over 90-minute intervals. The average of two consecutive measurements is used in equation (5) to calculate the radon source magnitude.

RESULTS

Survey of Radon Source Magnitude in U.S. Housing

Radon source magnitudes were determined in a survey comprising three housing groups: "energy-efficient" houses in the U.S. (one in Canada) (16 houses, Ho80); conventional houses in the San Francisco Bay Area (29 houses, Be79); and conventional houses in rural Maryland (56 houses, Mo81). Although this survey was not intended to be a random sampling, we believe the results provide an indication of what we might expect generally in the U.S. housing stock.

On the night prior to measurements occupants were asked to close windows and doors to establish a correspondence between the radon concentration measured and the ratio of source magnitude to air-exchange rate. Upon first entering the house, we collected one to three grab-samples of air for radon analysis; the samples were taken from the main living space, well away from doors and windows. A tracer-gas decay measurement of the air-exchange rate was then made over a one to two-hour interval. A summary of these measuremens is presented in Table 1.

In all three housing groups the air-exchange rates were found to be fairly low -- an arithmetic mean of 0.41 h^{-1}. In as much as all doors and windows were closed during the measurements, these values represent infiltration only (i.e., uncontrolled leakage through the building envelope). Furthermore, because most of the measurements were made in the spring and summer when the weather is mild, we can assume that the average annual infiltration rate for these houses is higher than the values we report.

A histogram (see Figure 3) of radon source magnitudes measured in all 101 houses shows a broad distribution of values, ranging from 0.01 to 6 pCi l^{-1} h^{-1}. Individual measurements are distributed in an approximately log-normal fashion, with a geometric mean of 0.37 pCi l^{-1} h^{-1}, and a geometric standard deviation of 4.0. The geometric means of the radon concentration and radon source magnitude measured in the San Francisco Bay Area houses were considerably lower than the corresponding values for the other two housing groups.

Time-Dependent Measurements of Radon Source Magnitude in Selected U.S. Houses

In the six houses located in the northeastern United States, we measured radon source magnitudes over periods of 3 to 30 days. Five of these houses, located in New York and Maryland, were built within the last ten years; all were tract houses, built with some attention to energy efficiency. The sixth house, located in New Jersey, is over 100 years old but has been retrofitted to reduce energy use. In three of the houses radon measurements were made continuously and air-exchange rate measurements were made intermittently, typically once per day. In the other three houses, both air-exchange rate and radon concentration were monitored continuously over three-to five-day periods.

Four of the houses, those in New York and New Jersey, were occupied during the measurements. Because of the cold weather, the windows and doors in all of the houses were closed almost all of the time. A summary of the results is presented in Table 2, which gives both the arithmetic mean and the range of the radon concentration, the air-exchange rate, and the radon source magnitude for each of the six houses. With the exception of Roch 49, which exhibits a much lower radon concentration and source magnitude, the mean values of each parameter obtained for these houses are narrowly distributed. The radon source magnitudes, especially in CB, AM-1 and SG-1, were found to vary significantly over the monitoring period, and the air-exchange rates were found to vary over a comparable or narrower range.

We observed a positive correlation between the measured air-exchange rate and radon source magnitude for five of the houses, as indicated in Figures 4 through 8. (In the case of Roch 49, the concentrations of radon were too low to determine whether or not such a correlation existed.) Figures 4 and 5 are scatter plots of the measurements made in CB and Roch 6, respectively, and the line plotted in each figure represents the least-squares fit to the data. For CB this line has a slope of 2.57 pCi l^{-1}, and a y-intercept of 0.23 pCi l^{-1} h^{-1}; the respective values for Roch 6 are 1.09 pCi l^{-1} and 0.21 pCi l^{-1} h^{-1}. Figures 6, 7, and 8 are plots of the air-exchange rate and radon source magnitude vs. time measured in Roch 60, AM-1 and SG-1, respectively. The average wind speed, derived from measurements made on-site at five-minute intervals, is also plotted for AM-1 (Ke81). In each of the three houses, the highest peaks in the radon source magnitude correspond in time with the highest peaks in the air-exchange rate. This correspondence is most evident in Figure 7, the plot for Am-1, showing that on 4/18 when the radon source magnitude increased from 0.7 to 2.6 and then decreased to 0.35 pCi l^{-1} h^{-1}, the air-exchange rate increased from 0.26 to 1.22 and then decreased to 0.72 h^{-1}.

DISCUSSION

Source Magnitude Distribution

Knowledge of the distribution of radon source magnitudes in U.S. housing would contribute greatly to efforts towards characterizing public exposure to indoor radon, as well as towards designing programs to reduce that exposure. Because our survey did not represent a random sampling of U.S. houses, the distribution of values we have reported may well differ from the actual distribution of radon source magnitudes. For example, half of the 16 energy-efficient houses surveyed were solar homes, generally relying on rock-bed heat storage, a potentially significant source of radon. The other two housing groups, although they reflect more typical design and construction practices, were selected from only two geographic areas of the country. Our data may also show a seasonal bias since most measurements were made in the spring and summer months; if, as we have observed, large variations in radon source magnitude occur over time, then it is very possible that systematic differences also occur from one season to another. Another item affecting the interpretation of these measurements is the contribution of radon in the outside air to the effective radon source magnitude. When trying to ascertain the effect of changes in the air-exchange rate on indoor radon concentrations, particularly for houses at the lower end of the distribution, this factor assumes importance.

We mention two other factors which, although potentially important, are unlikely to have much influence on the measured distribution: (1) the uncertainties associated wih the imprecision in measuring radon concentrations and the air-exchange rates as well as in approximating steady-state conditions, and (2) the apparent variability in radon source magnitude over short time periods within a given house. These two factors could cause a dispersion of the values reported here even if all of the houses had the same mean value; however, even this dispersion would not account for the wide range of source magnitudes observed in our survey. In other words, in spite of these considerations we conclude that the radon source magnitudes in U.S. houses are indeed widely distributed, and a substantial fraction are on the order of several pCi l^{-1} h^{-1}.

In most of the houses with source magnitudes on the order of 1 pCI l^{-1} h^{-1} or more, it is unlikely that building materials or domestic water are the dominant sources. Ingersoll measured radon emanation rates from 100 samples of concrete from across the United States and found values ranging from 0.2 to 2.0 pCi kg^{-1} h^{-1} (In81). If we take the case of a one-story house with 2.4 m ceilings, then a 0.2 m-thick concrete slab with a typical density of 2000 kg m^{-3} would contribute only 0.02 to 0.2 pCi l^{-1} h^{-1} to the source magnitude. We took a few measurements of radon in tap water in Maryland and found concentrations on the order of 1000 pCi/l; using Hess' observation that 10,000 pCi/l of radon in water typically results in 0.65 pCi/l of radon in air in a house with an air-exchange rate of 1 h^{-1}, the resulting source magnitude would be 0.07 pCi l^{-1} h^{-1}. On the other hand, Wilkening reports that data from roughly 1000 measurements of radon flux from soil range up to 1.4 pCi m^{-2} s^{-2} (mean value of 0.4 pCi m^{-2} s^{-2}), a value which would contribute 2.1 pCi l^{-1} h^{-1} (mean value of 0.7 pCi l^{-1} h^{-1}) to the source magnitude in the house postulated above, assuming the flux from the soil into the house is the same as the flux from uncovered soil (Wi72). The fact that even the maximum flux from uncovered soil is not large enough to account for the highest indoor source magnitudes observed suggests the possibility that in some cases radon may be more efficiently transported from soil into a house than into the atmosphere.

In addition to the variations observed from one house to another our survey revealed a significant difference in radon source magnitude between the San Francisco Bay Area and the communities studied in rural Maryland. This difference suggests that radon source magnitude may depend on geological or structural factors. If so, it may be useful to conduct limited surveys of radon source magnitudes in a region to determine whether radon levels are high enough to justify further measurements in that area.

That radon source magnitudes show a broad log-normal distribution has important implications for any effort to control public exposure to radon in houses. For such a distribution, the best cost-benefit ratio can be obtained by addressing control efforts toward those houses with the highest source magnitudes. Since the source magnitude in closed houses appears to be more broadly distributed than the air-exchange rate, specifying a minimum air-exchange rate for all houses does not appear to be a cost-effective control strategy.

Correlation Between Air-Exchange Rates and Radon Source Magnitude

The correlation between air-exchange rate and radon source magnitude is evident in Figures 4-8. The correlation is particularly clear for the two houses with the highest (mean) source magnitudes, CB and AM-1. It is our hypothesis that in these two houses, and perhaps in Roch 6, Roch 60 and SG-1, as well, surrounding soil is the dominant source of radon; the radon enters the house in high concentration with soil gas that is driven by the same forces that cause infiltration -- wind speed and indoor-outdoor temperature difference. A simple model consistent with this correlation is that on average a constant fraction of the air that infiltrates the house comes from the soil and carries with it a high concentration of radon; as the wind speed and temperature difference increase, the flow rate of soil gas into the house also increases, therefore, the radon source magnitude increases. For example, if the radon concentration in the soil gas is 500 pCi/l, a commonly reported value, then the soil gas portion of infiltration air need be only 0.5% of the total (corresponding to 16 l/m at an air-exchange rate of 0.4 h^{-1}) to account for the slope of the line in Figure 4. The y-intercept of 0.2 pCi l^{-1} h^{-1} could represent the contribution to the source magnitude of radon diffusing from and through the building materials. Much of the scatter in these data could reflect changes in the concentration of radon in the soil gas at the soil/house interface, or changes in the fraction of infiltrating air coming from the soil.

The data for AM-1 also show a strong correspondence between radon source magnitude and air-exchange rate, particularly on 4/18 when the air-exchange rate steadily rose over a period of 20 hours, increasing greatly during the middle of this period concomitantly with an increase in wind speed. During the first 15 hours of this period, the radon source magnitude also increased strongly; however at noon, when the last significant increase in air-exchange rate (to 1.2 h^{-1}) occurred, the radon source magnitude declined by nearly 25%. In fact, throughout this period it appears that the increases in radon source magnitude become smaller relative to the increases in the air-exchange rate. It is possible that the concentration of radon in the soil gas drops as the soil gas is drawn into the house at a higher rate. That the source magnitude dropped to a low value of 0.35 pCi l^{-1} h^{-1} that afternoon, even though the air-exchange rate was still fairly high at 0.7 h^{-1}, is consistent with this hypothesis.

The correlations we observed between air-exchange rate and radon source magnitude would not be expected to apply if the changes in air-exchange rate were a result of opening windows and doors or changing the rate of mechanical ventilation. As pointed out earlier, measurements in these houses were made during winter and spring when the outside temperatures are relatively low, and therefore, we assume that the doors and windows were closed as much as possible. Given these conditions the changes in air-exchange rates can be assumed to reflect changes in weather conditions which appear also to effect the radon source magnitude in some of these houses.

We have considered various explanations for the observed correlation between radon source magnitude and air-exchange rate, but in the case of two houses, CB and AM-1, the data seem to point quite clearly to the aforementioned soil-gas theory. For example, since we include in our calculation of source magnitude the contribution from outdoor radon, we expect a positive correlation between the source magnitude and the air-exchange rate; however the observed slopes are much steeper than the slope of the regression line resulting from this effect alone, which would equal the outdoor radon concentration, typically 0.1 pCi/l. Errors in measuring the air-exchange rate could also lead to a false correlation. For example, if the radon concentration and air-exchange rate were constant, then a scatter plot of a series of measurements of radon source magnitude and air-exchange rate would show a positive correlation with a slope equal to the radon concentration. The range of air-exchange rates in such a plot would reflect the precision of the measurement. How precise our air-exchange rate measurements are is difficult to estimate and probably varies from one house to another, depending on the mixing rates within the house and on the number and location of sampling lines. The first eight measurements in Roch 60 indicate that it is possible to achieve a standard deviation of less than 0.05 h^{-1} in the measurement of air-exchange rate. Such a level of precision is sufficient to preclude the possibility that the correlation between air-exchange rate and radon source magnitude for CB and AM-1 is due to measurement uncertainty. In the case of Roch 6, Roch 60 and SG-1 however, the apparent correlations could reflect measurement uncertainty in as much as the measurements of air-exchange rate in each of these houses are distributed over fairly narrow ranges.

CONCLUSIONS

We have measured radon source magnitudes in over 100 houses by simultaneously measuring the radon concentration and the air-exchange rate; in six of these houses source magnitude measurements were made over time, in some cases continuously. The data from these measurements corroborate the hypothesis that soil is an important source of indoor radon in U.S. houses, and, furthermore, support the theory that soil gas influx, rather than molecular diffusion, is the dominant transport process by which radon from soil enters houses, at least in those with large source magnitudes.

Such studies could be fruitfully extended in at least two directions: A well-designed survey project could be undertaken to determine the actual distribution of radon source magnitudes in the U.S. housing stock to provide a basis for regulatory action aimed at controlling public exposures to radon. Continuous measurement of the source magnitude over a period of three months to a year in individual houses where source magnitudes are high would also be useful, especially if measurements of the radon concentration in soil gas, the radon flux through an opening between the house and the soil, and weather parameters were made simultaneously.

ACKNOWLEDGEMENTS

We thank the following people for their assistance in collecting and analyzing the data on radon concentrations and air-exchange rates for the six houses monitored over time: M. Lints of the Rochester Institute of Technology, and R. Young, S. Brown, J. Dillworth, J. Pepper, F. Offermann, S. Doyle, and B. Moed, all of Lawrence Berkeley Laboratory.

This work was supported by the Assistant Secretary for Conservation and Solar Energy, Office of Building and Community Systems, Buildings Division, and the Assistant Secretary for Environment, Office of Health and Environmental Research, Human Health and Assessments Division of the U.S. Department of Energy under Contract No. W-7405-ENG-48.

References

Be79 Berk J.V., Boegel M.L., Ingersoll J.B., Nazaroff W.W., Stitt B.D., and Zapalac G.H., 1979, "Radon Measurements and Emanation Studies," in *Energy Efficient Buildings Program: Chapter from the Energy and Environment Division Annual Report 1979*, Lawrence Berkeley Laboratory report, LBL-10704, Berkeley, CA.

Ge81 George A.C. and Fisenne I.M., 1981, private communication, Environmental Measurements Laboratory, New York, May 12.

He81 Hess C.T., Weiffenbach C.V., Norton S.A., Brutsaert W.F., and Hess A.L., 1981, "Radon-222 in Potable Water Supplies in Maine: the Geology, Hydrology, Physics, and Health Effects," presented at the Second Special Symposium on Natural Radiation Environment, Bhabha Atomic Research Centre, Bombay, India, January 19-23, 1981.

Ho80 Hollowell C.D., Berk J.V., Boegel M.L., Ingersoll J.G., Krinkel D.L., and Nazaroff W.W., 1980, *Radon in Energy-Efficient Residences*, Lawrence Berkeley Laboratory report, LBL-9560, Berkeley, CA.

In80 Ingersoll J.G., 1980, *Operating Instructions for LBL Radon Measurement Facilities*, Lawrence Berkeley Laboratory report, LBL-11097, Berkeley CA.

In81 Ingersoll J.G., 1981, "A Survey of Radionuclide Contents and Radon Emanation Rates in Building Materials Used in the United States," Lawrence Berkeley Laboratory report, LBL-11771, Berkeley, CA. Submitted to *Health Phys*.

Ke81 Kelly C.J., 1981, private communication, Automation Industries, Inc., Vitro Laboratories Division, Silver Spring, MD, May 12.

Lu57 Lucas H.F., 1957, "Improved Low-Level Alpha Scintillation Counter for Radon," *Rev.Sci Instrum. 28*, 680-683.

Lu77 Lucas H.F., 1977, "Alpha Scintillation Radon Counting," in *Workshop on Methods for Measuring Radiation in and around Uranium Mills* Albuquerque (E.D. Harwood, ed.), Vol. 3, No. 9, Atomic Industrial Forum, Washington, D.C.

Mo81 Moschandreas D.J., Rector H.E., and Tierney P.O., 1981, *A Survey Study of Residential Radon Levels*, Geomet Technologies report, ES-877, Rockville, MD.

Na81 Nazaroff W.W., Offermann F.J., and Robb A.W., 1981, "Automated System for Measuring Air-Exchange Rate and Radon Concentration in Houses," Lawrence Berkeley Laboratory report, LBL-12945, Berkeley, CA. Submitted to *Health Phys*.

Wi72 Wilkening M.H., Clements W.E., and Stanley D., 1972, "Radon-222 Flux Measurements in Widely Separated Areas," Natural Radiation Environment II (J.A.S. Adams et al, eds.), U.S. Energy Research and Development Administration report, CONF 26-720805, Washington, D.C.

Figure Captions

1. Schematic diagram of the Continuous Radon Monitor (CRM).

2. Schematic diagram of the mechanical system of the Aardvark, an automated system for continuously measuring air-exchange rate and radon concentration in occupied houses.

3. Frequency distribution of radon source magnitude. For each house, the radon entry rate per unit house volume is calculated as the product of the radon concentration and the air-exchange rate, measured after the house had been closed for several hours.

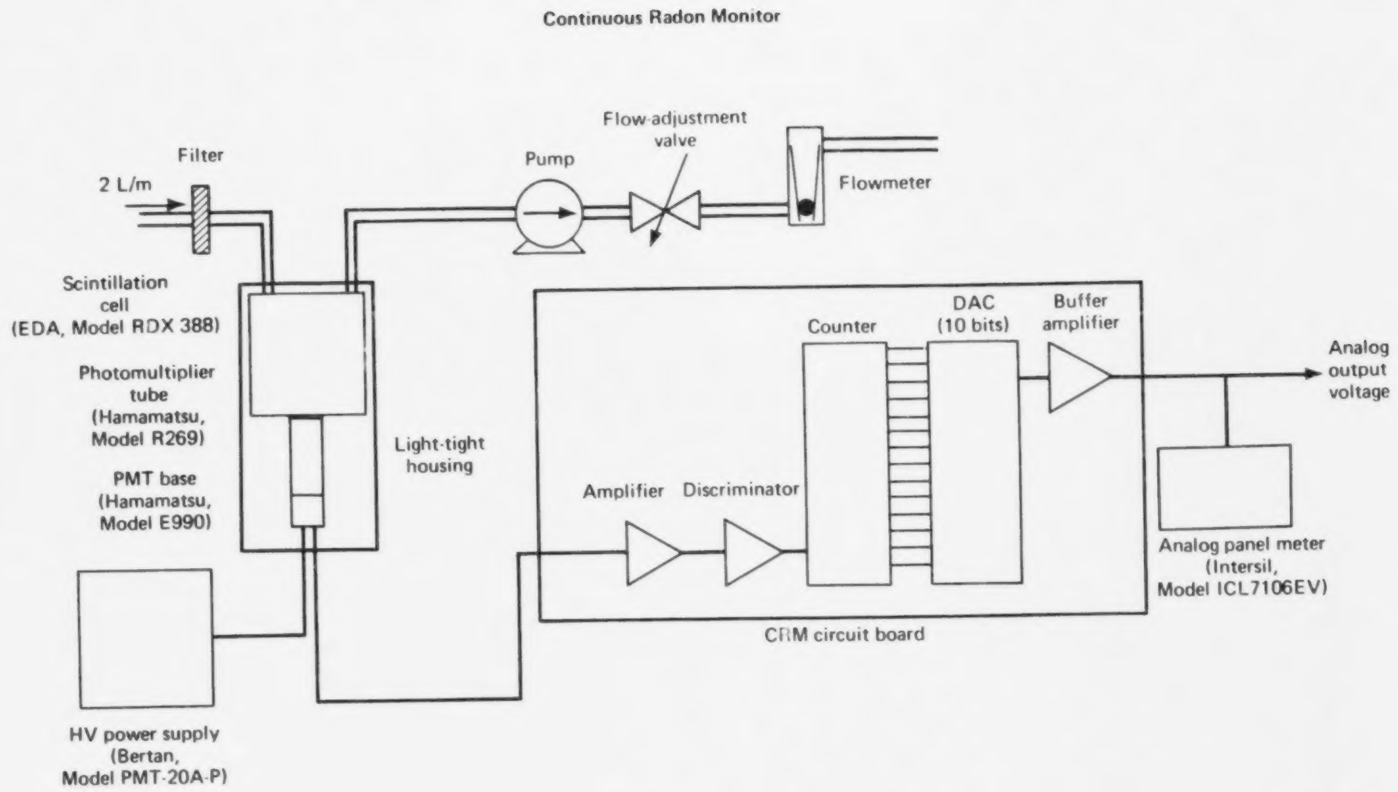
4. Scatter plot of radon source magnitude versus air-exchange rate for house, CB. The data were collected between November 16 and December 17, 1980. The line represents a least-squares fit to the data.

5. Scatter plot of radon source magnitude versus air-exchange rate for house, Roch 6. The data were collected between January 8 and January 22, 1981. The line represents a least-squares fit to the data.

6. Plot of air-exchange rate and radon source magnitude versus time for house, Roch 60.

7. Plot of wind speed, air-exchange rate, and radon source magnitude versus time for house, AM-1.

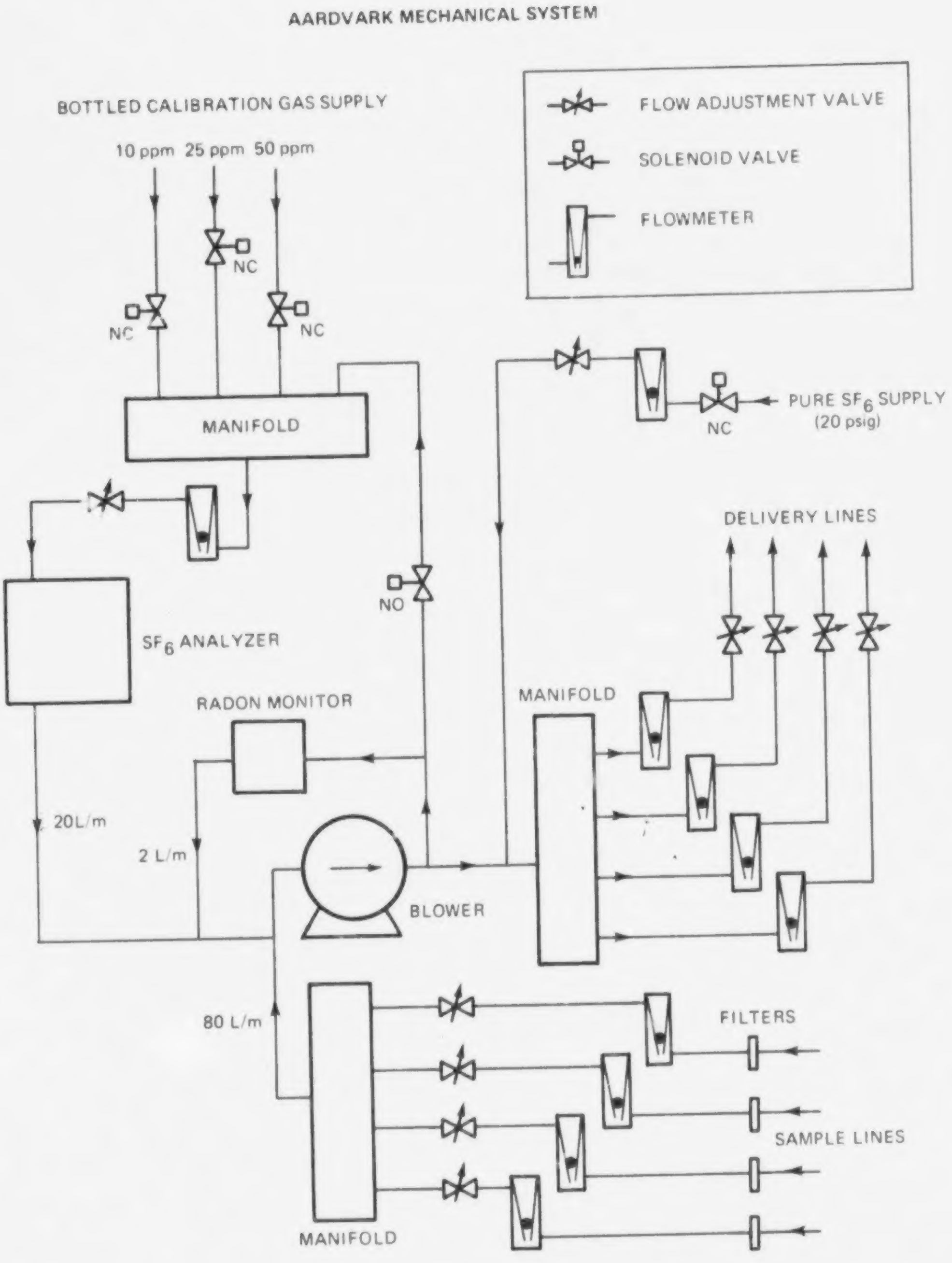
8. Plot of air-exchange rate and radon source magnitude versus time for house, SG-1.


Continuous Radon Monitor

2 L/m
Filter
Pump
Flow-adjustment valve
Flowmeter
Scintillation cell (EDA, Model RDX 388)
Photomultiplier tube (Hamamatsu, Model R269)
PMT base (Hamamatsu, Model E990)
Light-tight housing
HV power supply (Bertan, Model PMT-20A-P)
Amplifier
Discriminator
Counter
DAC (10 bits)
Buffer amplifier
Analog output voltage
Analog panel meter (Intersil, Model ICL7106EV)
CRM circuit board

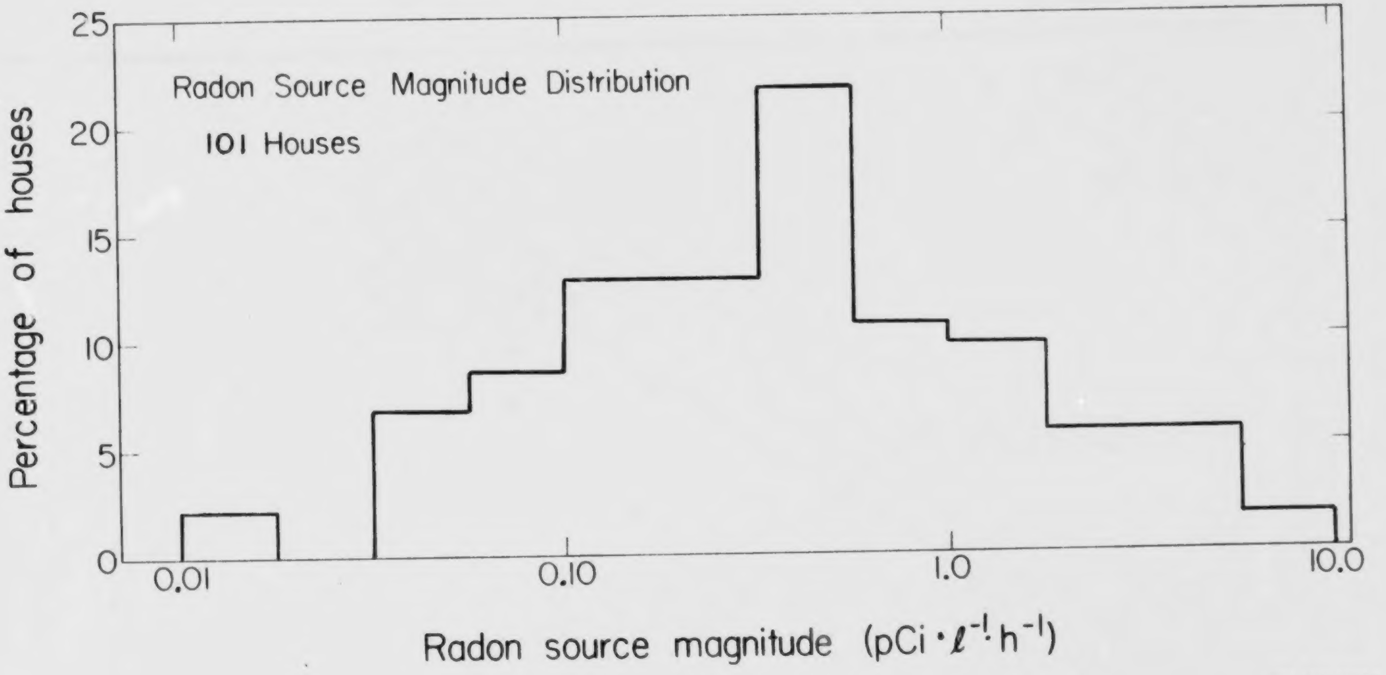
XBL 818-1070

Figure 1



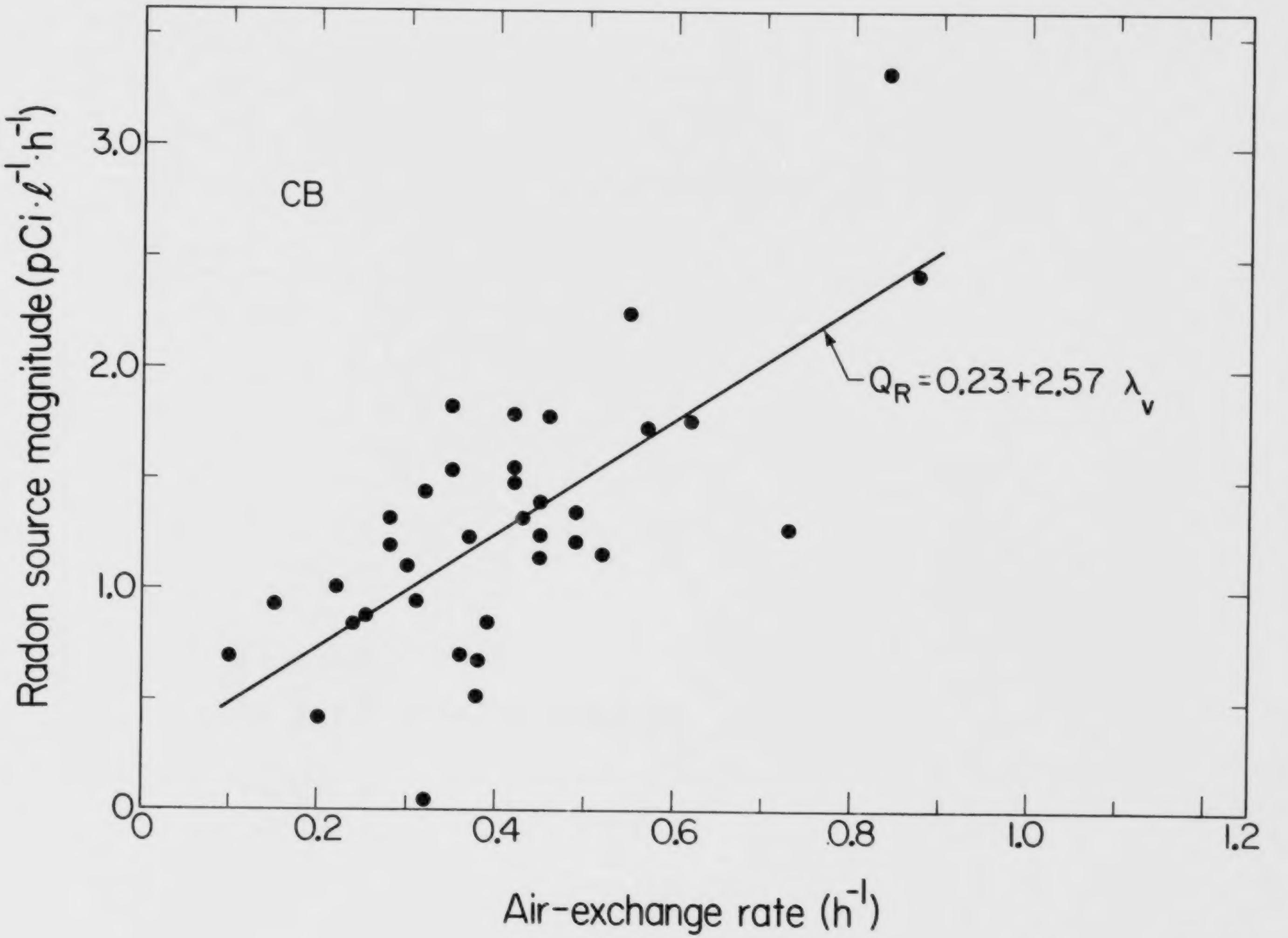
XBL 817-1036

Figure 2



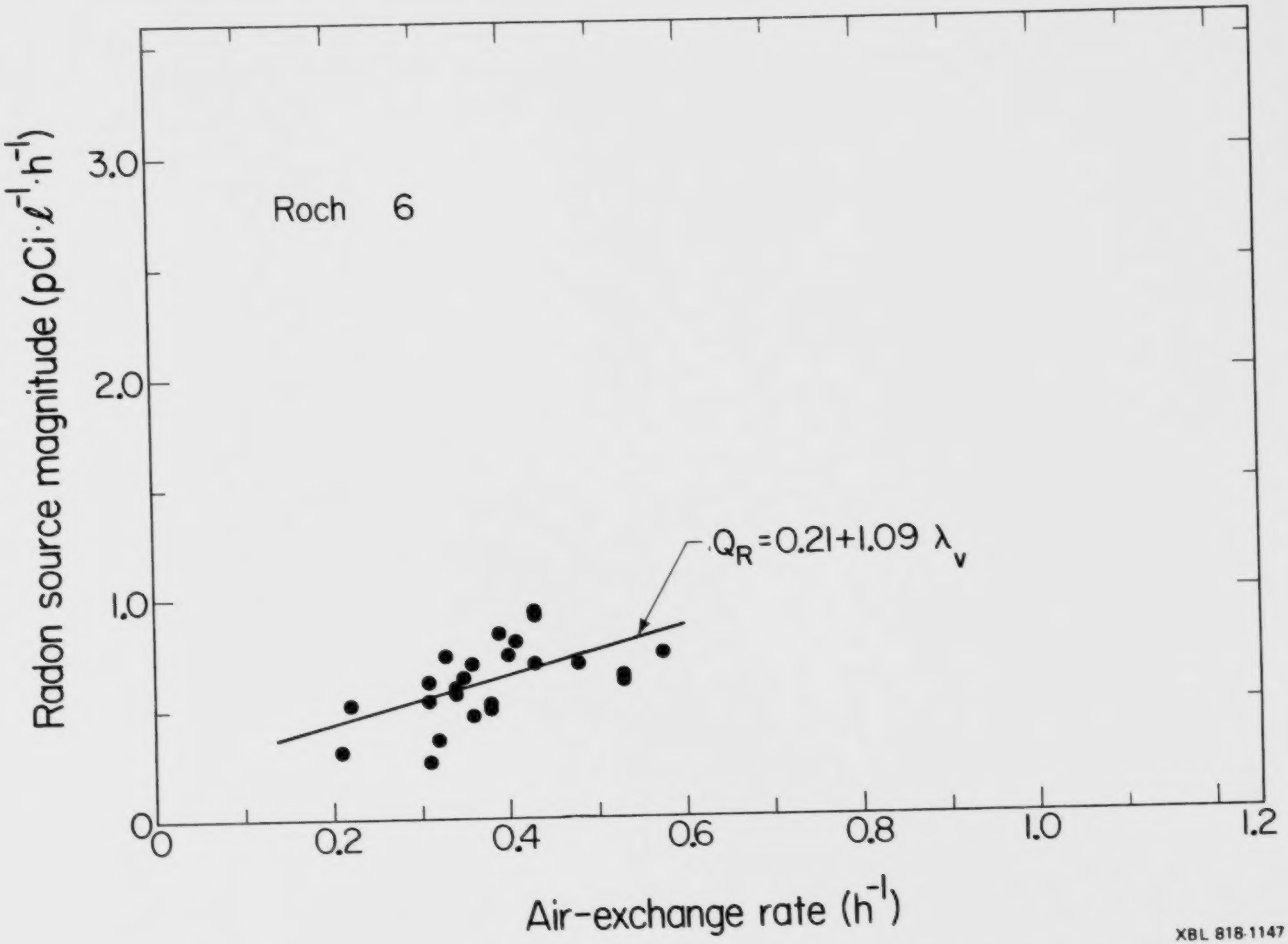
XBL 818-1146A

Figure 3



XBL 818-1148

Figure 4



XBL 818-1147

Figure 5

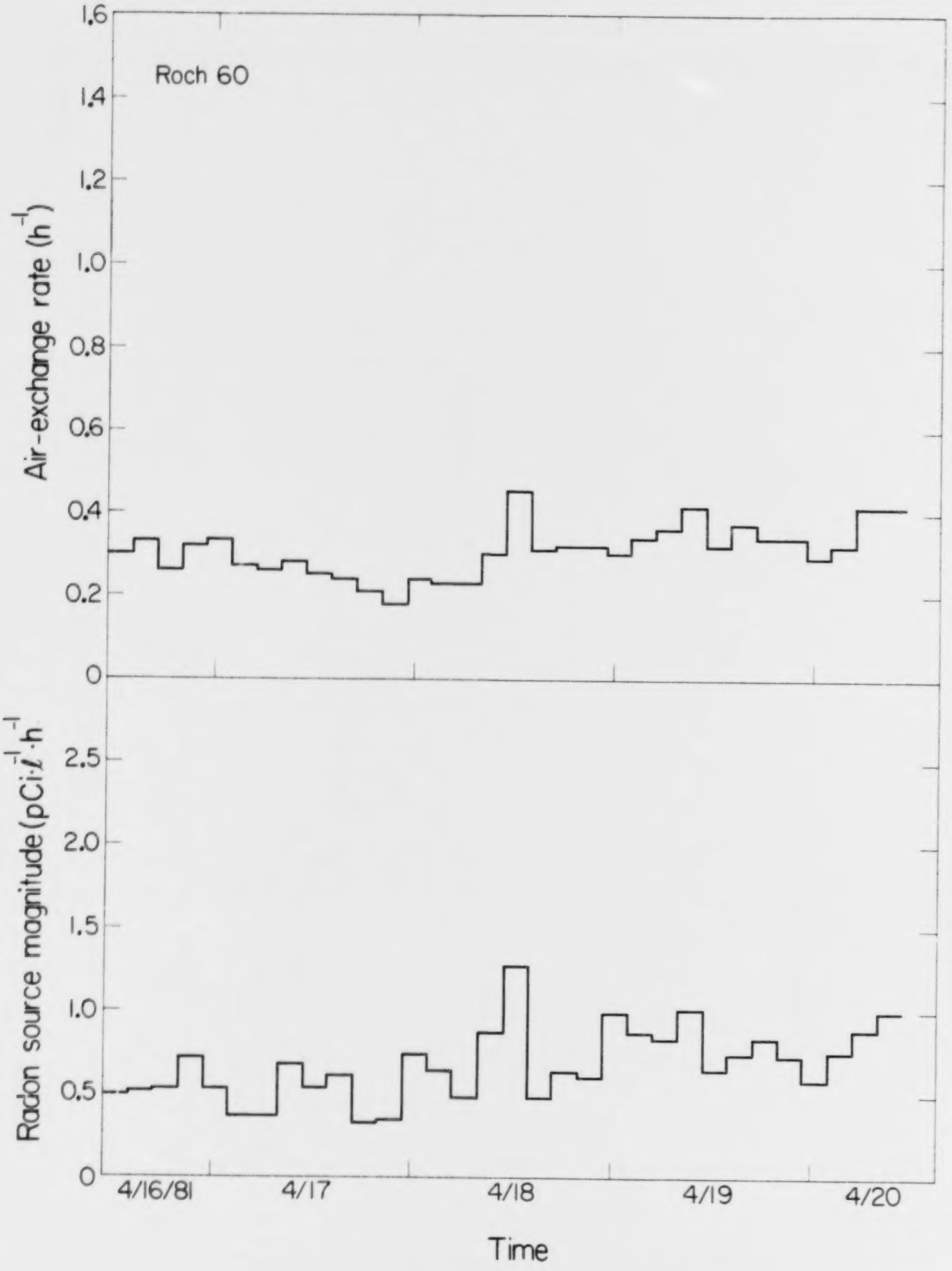
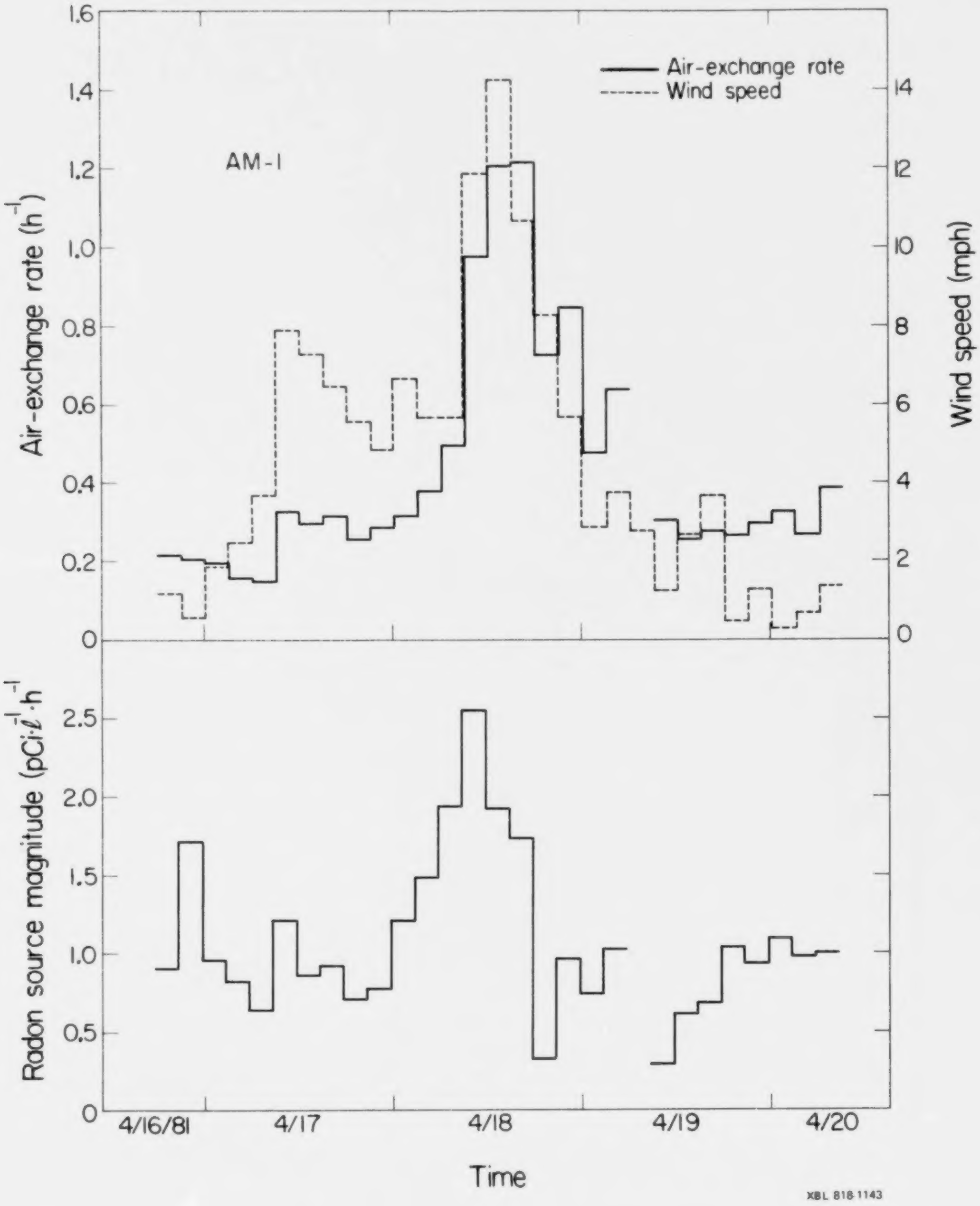

Roch 60
Air-exchange rate (h^{-1})
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0
Radon source magnitude (pCi·ℓ$^{-1}$·h^{-1})
2.5
2.0
1.5
1.0
0.5
0
4/16/81
4/17
4/18
4/19
4/20
Time
XBL 818-1145

Figure 6



Air-exchange rate
Wind speed
AM-1
Air-exchange rate (h^{-1})
Wind speed (mph)
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0
14
12
10
8
6
4
2
0
Radon source magnitude ($pCi \cdot \ell^{-1} \cdot h^{-1}$)
2.5
2.0
1.5
1.0
0.5
0
4/16/81
4/17
4/18
4/19
4/20
Time
XBL 818-1143

Figure 7

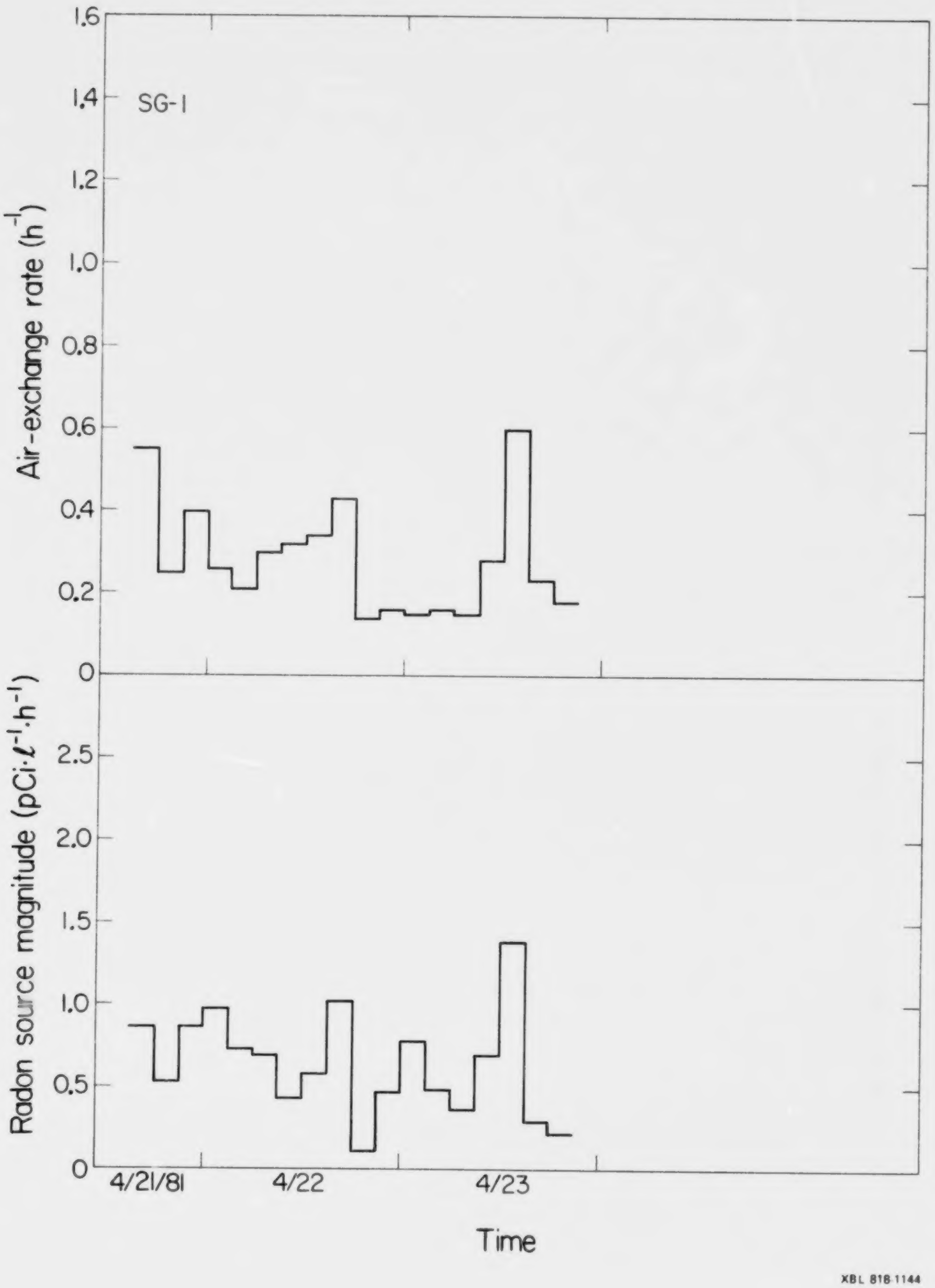

SG-1
Air-exchange rate (h^{-1})
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0
Radon source magnitude (pCi·ℓ$^{-1}$·h^{-1})
2.5
2.0
1.5
1.0
0.5
0
4/21/81
4/22
4/23
Time
XBL 818-1144

Figure 8

Table 1. Data from 101-House Survey of Radon Concentration and Air-Exchange Rate

Sample	Monitoring Period	No. of Houses	Radon (pCi/1) GM[a]	Radon (pCi/1) GSD[b]	Air-Exchange Rate (h^{-1}) GM[a]	Air-Exchange Rate (h^{-1}) GSD[b]	Source Magnitude (pCi 1^{-1} h^{-1}) GM[a]	Source Magnitude (pCi 1^{-1} h^{-1}) GSD[b]
U.S. Energy-Efficient Residences[c]	May-August 1979	16	2.6	2.2	0.23	2.2	0.61	2.5
San Francisco Bay Area[d]	July-September 1979	29	0.4	2.2	0.28	2.5	0.10	2.5
Maryland[e,f]	May-October 1980	56	1.8	4.0	0.35	2.2	0.62	3.4
Total		101	1.2	4.0	0.31	2.3	0.37	4.0

[a] $GM = \exp\left[\frac{1}{N} \sum_{i=1}^{N} \ln X_i\right]$

[b] $GSD = \exp\left[\frac{\sum_{i=1}^{N} [\ln X_i - \ln(GM)]^2}{(N-1)}\right]^{\frac{1}{2}}$

[c] Source: Hollowell, C.D., Berk, J.V., Boegel, M.L., Ingersoll, J.G., Krinkel, D.L., and Nazaroff, W.W., 1980, Radon in Energy-Efficient Residences, Lawrence Berkeley Laboratory Report LBL-9560, Berkeley, CA.

[d] Source: Berk, J.V., Boegel, M.L., Ingersoll, J.G., Nazaroff, W.W., Stitt, B.D., and Zapalac, G.H., 1979, "Radon Measurements and Emanation Studies," in Energy Efficient Buildings Program: chapter from the Energy and Environment Division Annual Report 1979, Lawrence Berkeley Laboratory Report LBL-10704, Berkeley, CA.

[e] Source: Moschandreas, D.J., Rector, H.E., and Tierney, P.O., 1981, A Survey Study of Residential Radon Levels, Geomet Technologies Report ES-877, Rockville, MD.

[f] In six of these 56 houses, the radon concentration was found to be less than the detection limit of the measurement procedure (0.4 pCi/1 with 50% relative standard deviation). For these houses the radon concentration was assumed to be 0.1 pCi/1 for purposes of calculating the geometric mean radon concentration and source magnitude.

Table 2. Summary of Radon Source Magnitude Measurements for Six Houses Monitored Over Time

House ID	Location	Monitoring Period	No. of Meas.	Radon[a] (pCi/l)	Air-Exchange[a] Rate (h^{-1})	Radon Source Magnitude[a,e] (pCi l^{-1} h^{-1})
CB[b]	N.J.	11/16- 12/17/80	36	3.2 (0.7-5.1)	0.41 (0.1-0.87)	1.3 (0.0-3.3)
ROCH 6[b]	N.Y.	1/8- 1/22/81	25	1.6 (0.8-2.2)	0.38 (0.21-0.55)	0.65 (0.3-0.9)
ROCH 49[b]	N.Y.	2/6- 2/20/81	11	0.1 (0.0-0.4)	0.40 (0.21-0.57)	0.05 (0.0-0.2)
ROCH 60[c]	N.Y.	4/16 4/20/81	32	2.2 (1.2-3.0)	0.31 (0.18-0.45)	0.7 (0.4-1.3)
AM-1[d]	MD.	4/16- 4/20/81	28	2.9 (0.6-5.5)	0.44 (0.15-1.22)	1.1 (0.3-2.6)
SG-1[d]	MD.	4/21- 4/23/81	18	2.3 (1.4-3.3)	0.28 (0.14-0.60)	0.65 (0.1-1.4)

[a] Arithmetic mean values for three-hour intervals; range of values given in parentheses.

[b] Occupied house, radon measured by CRM, air-exchange rates measured by tracer gas decay one or two times per day.

[c] Occupied house, radon measured by CRM, air-exchange rates measured continuously by tracer gas decay.

[d] Unoccupied house, radon measured by CRM, air-exchange rate measured continuously by tracer decay.

[e] The mean radon source magnitude is rounded to the nearest 0.05 pCi l^{-1} h^{-1}.

FIELD AND LABORATORY MEASUREMENTS OF RADON CONCENTRATIONS IN MAINE HOUSES

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University of Texas, Houston, Texas

ABSTRACT

Radon measurements using track etch cups and diffusion alpha detectors in the field are compared with air grab samples and water grab samples measured in the laboratory for 100 houses in Maine. These measurements have been taken for the winter months from October 1980 to April 1981 as part of an on-going year round study. Airborne radon concentrations ranging from 0.05 to 210 pCi/l have been measured. Radon in water concentrations from 20 to 180,000 pCi/l have been measured. A linear regression radon concentration in air versus radon concentration in water is significant at the r = 0.5 significance level for 40 laboratory samples of each. The concentration of radon in air is 1.3 pCi/l for 10,000 pCi/l in water. Analysis of constant sources of radon in air such as soil, bedrock, or building materials and variable sources of radon such as radon rich water shows a strong inverse effect on concentration due to building ventilation.

There has been great interest in indoor radon in houses in recent publications by Barnaby on radon in Swedish houses, (Ba 79), by Cliff on radon in dwellings in Great Britain (Cl 78, Cl 79), by Fleisher in Rochester, New York (Fl 81), and by the U.S.E.P.A. in houses built on phosphate tailings piles in Florida (US 75). Recommendations for changes have been made for radon levels above 3 pCi/l in homes. In Canada recommendations for changes have been made for radon greater than 5 pCi/l, in places such as Uranium City, Saskatchewan (Ke 78).

This paper presents the initial results of a cooperative project between the University of Maine Department of Physics, and the University of Texas School of Public Health to measure indoor air and water radon levels in 100 houses in Maine. The radon in air was measured in the laboratory with using 96 grab samples and with 514 track etch cups. The radon in water was measured in the laboratory using 100 grab samples from the same houses. The houses were also sampled for uranium in water and were characterized for fuel used, estimate of ventilation, presence of granite building materials, and with types of construction, number of residents, and volume of house. The survey took place in October, 1980, through May, 1981, and is part of a year round study of radon level in houses. The initial grab samples, of air and water, and the initial place of Terradex - F1 track etch cups with paper filters took place in October, 1980, and the cups were collected in April - May, 1981, and then analyzed by Terradex Corp. The data thus represents radon levels during fall and winter months, when the ventilation is at a minimum in Maine. The track etch cups were placed in bedroom, bathroom, living room or kitchen, basement, and outside on a porch. The grab sample of 5 liters of air was collected in Tedlar or heavy plastic bags, in the kitchen or living room of a house, while students were collecting the information about the house. The water sample was then taken after the water was allowed to flow in a sink for at least 10 minutes to permit fresh water to be brought up out of the well. The 10 ml water sample was taken with a syringe and placed in liquid scintillation vials with 10 ml mineral oil fluor and analyzed for 40 minutes with a commercial liquid scintillation counter. Samples were standardized with radium solutions; for further details see the method of Prichard and Gesell (Pr 80). A 10 liter water sample was taken and acidified with 10 ml 1 normal HCl for radium or uranium analysis. Track etch cups were provided by the U.S.E.P.A. Laboratory in Las Vegas, Nevada and analyzed with low sensitivity and high sensitivity depending on the density of tracks. After the cups were exposed for 6 months - 7 months, the cups were collected, sealed together with plastic inserts and stored in Tedlar bags until returning them to be measured at Terradex Corp. in California. Cups were identified by track etch cup number and house code number.

The houses measured were taken from a group of houses previously studied for radon in water. The houses were selected for a high radon in water group and a low radon in water group. Three fourths of the houses were measured by student teams and one fourth by the State of Maine Department of Health Engineering. All the cups were handled in the same way and the survey is being continued this summer using multiple cups at the same point in the house. Of the original 530 cups, 513 have been used and measured, a return of more than 95% of cups placed.

Radon in air was measured in the grab samples by extracting the radon in -70°C Hexane, and counting it with commercial liquid scintillation counters. Additional data were collected in several houses using electronic diffusion alpha detectors of the Wrenn type (Wr 75). The diffusion alpha detector was calibrated using National Bureau of Standards radium sources and recalibrated at the Eastern Environmental Measurements Laboratory, and other university laboratories, at the University of Texas and University of Maine, and U.S.E.P.A. lab at Montgomery, Alabama. The alpha measurements of radon were taken over a period of two days to 7 days at 10 minute intervals. These measurements were used to determine the variation of radon with time in a variety of houses with various sources of radon in air. The data was analyzed for peaks and compared to records of water use and changes in ventilation. Analysis of pulse shapes permitted determination of ventilation times for some of the houses.

RESULTS

The radon in air values for grab samples of air ranged from .03 to 201 pCi/l while the track etch cups ranged from 0.06 to 132 pCi/l. These values are illustrated in Figure 1 which compares the grab sample taken in October with the average value for the track etch cup taken from October through May. The data are correlated significantly with r = 0.991, with a slope of 1.49 and an intercept of -0.48 pCi/l. This result may show a systematic measurement difference or may show a difference in average ventilation. To test average radon levels due to radon from water, which ranged from 20 to 115,000 pCi/l, plots were made of bathroom air measured with track etch cups versus radon in water concentration. These results are shown in Figure 2. This data has a slope of 0.107 and an intercept of 1.59 with r = .307, n = 85 significance. A similar curve was made for radon in the basement air measured by track etch cup and water radon concentration in Figure 3. The data has a slope of 0.127 and an intercept of 4.49 with r = 0.151 significance. A summary of all data for track etch cups versus water radon is shown in Table I. Surprisingly, the highest value for slope is for radon in basement air versus radon in water. Significant lower slopes are shown for all bathroom and kitchen-living room.

Histograms number of occurrences versus radon levels by room, measured using track etch cups are shown in Figure 4. The average values for radon in air are shown with arrows and are as follows; bathrooms 3.51 ± 7.12, bedrooms 3.10 ± 6.98, kitchen-living rooms 3.21 ± 8.27, basement 6.79 ± 16.8, outside 0.83 ± .79. Since the radon is highest in basements, this shows that a major source is associated with soil gas, or water radon in floor drains, or basement building materials. Reductions on the second floor are by 2, and reductions on third floors are also observed to be factors of 2 from two story house data. Error estimates are taken for the data to simulate a normal distribution.

By using calibrated Spitz-Wrenn meters (Wr 75), measurements have been made of levels of radon in air during water uses such as showering, dish washing, and clothes washing. The fraction of radon lost ranges from 50% to 95% for these activities (Pa 79). Weekly averages of the radon in the 20 houses showed radon levels from 0.5 pCi/l to 87 pCi/l during the heating season. Occasional radon daughter measurements by the method of Kusnetz (Ku 56) showed equilibrium of daughters from 0.3 to 0.5. Portions of records of radon concentration in air of three houses A, B and 18 are shown in Figures 5 and 6. House A has the highest levels we have found. This house, with an average of 87 pCi/l is a solar heated house with connected greenhouse built on granite bedrock with 2 feet of granite gravel fill. House B shows no ^{222}Rn contribution other than water supply. These results show that the radon in water is not always the most significant source of radon in the air; but that radon in water usually high when radon from all sources is high. Other sources include granite blocks in basement walls or granite gravels in concrete or direct emanation into houses from granite through soil and foundations.

Results exhibiting the variation of radon in air in homes with the radon in the water are summarized in Figure 7. The open circles represent average radon in the air due to water supply only obtained by considering only the radon appearing during times of major water uses and which decreased with time according to apparent ventilation rates of the houses studied - see Figure 5. For the component of radon due to water, the radon levels in the air have been normalized to these levels expected for a ventilation rate of one air change per hour. The unadjusted average radon levels are also plotted as asterisks for all the houses studied. All data presented are for winter, with minimum ventilation in the houses. Since summer ventilation decreases levels in houses, the expected exposures from radon in these sixteen houses range from 0.01 to 14 working level months per year.

TABLE I

	Slope	Intercept	Signif.	No. Pts.	No.≥3 pCi/l	Mean	S.D.
Bathroom	0.107	1.59	0.307	85	23	3.39	6.9
Kitchen/Living Room	0.106	1.58	0.216	71	15	3.11	7.9
Bedroom	0.069	1.81	0.198	84	17	2.98	6.7
Basement	0.127	4.49	0.151	79	38	6.44	16.1
Outside	0.0085	0.710	.218	68	2	0.84	.78

References

Ba79 Barnaby, W. 1979, "Very high radiation levels found in Swedish houses." Nature 281, 6.

Cl 78 Cliff, K.D. 1978, "Assessment of airborne radon daughter concentrations in dwellings in Great Britain." Phys. Med. Biol. 23, 696.

Cl 79 Cliff, K.D., Davies, B.C. and Reissland, J.A. 1979, "Little danger from radon." Nature 279, 12.

Fl 81 Fleischer, R.L., Mongro-Campero, A. and Turner, C.G., 1981, "Radon levels in homes in the Northeastern United States." Second Special Symposium on Natural Radiation Environment Bhabha Atomic Research Center, Bombay, India.

US 75 United States Environmental Protection Agency (1975). Preliminary findings: radon daughter levels in structures constructed on reclaimed Florida phosphate land. Technical note, EPA ORP/CSD 75-4.

Ge 79 George, A.C. and Breslin, A.J., 1979, "Distribution of ambient radon and radon daughters in New York - New Jersey residences." Paper in Proc. Nat. Rad. Environ III Edited by Gesell, T.F., Lowder, W.M. and McLaughlin, J.E. Con. 780422 (Dept. of Energy, Washington).

Ke 78 Keith Consulting, 1978, Report on investigative and remedial measures, radiation reduction and radioactive decontamination in Uranium City, Saskatchewan for the Atomic Energy Control Board of Canada. Keith Consulting of Regina, Prince Albert, Lethbridge, Edmonton, Canada.

Pa 79 Partridge, J.E., Horton, T.R., and Sensintaffar, E.L., 1979, "A study of radon -222 released from water during typical household activities" U.S.E.P.A. Technical note ORP/EERF-79-1 Eastern Environmental Radiation Facility, Montgomery, Alabama 36109.

Pr 77 Prichard, H.M. and Gesell, T.F., 1977, "Rapid measurements of ^{222}Rn concentrations in water with a commercial liquid scintillation counter." Health Physics 33 No. 6, 577-581.

Pr 81 Prichard, H.M., 1981 "The measurement of radon-222 in air by direct extraction into a liquid scintillator." International Meeting on radon-radon daughter measurements, Montgomery, Alabama.

Wr 75 Wrenn, M.D., Spitz, H. and Cohen, N., 1975, "Design of a continuous digital output environmental radon monitor." IEEE Transactions of Nuclear Science N.E. -33, 645.

Ku 56 Kusnets, H.L., 1956, "Radon daughters in mine atmospheres: A field method for determining concentrations." AM. IND. HYG. Q. 17(1): 85-88.

FIGURE CAPTIONS

Figure 1. Radon concentration in air pCi/1 measured by track etch cups from October, 1980 to May, 1981 and grab samples of air taken in October, 1980.

Figure 2. Radon concentration in bathroom air measured by track etch cups versus radon in water measured by liquid scintillation.

Figure 3. Radon concentration in basement air measured by track etch cups versus radon in water measured by liquid scintillation.

Figure 4. Histograms of number of occurrences of radon levels by room measured with track etch cups versus concentration of levels in the room for outside air, kitchen/living room air, bedroom air, bathroom air and basement air. Average values indicated by arrows.

Figure 5. Radon concentration in air for two different houses measured by diffusion alpha detectors at 10 minute intervals versus time of day: a) open window to outside; b) close window to outside; c) run clothes washer; d) shower; e) partially close off greenhouse from house; f and g) showers; h) dishwasher; i) bath and shower. Note for houses the effect of individual water uses in comparison to background ^{222}Rn.

Figure 6. Radon concentration in House 18 versus time of day for 6 days, noon marked with n. Showers are marked with letter s, baths with letter b, laundry with letter l and dishwasher marked with letter d.

Figure 7. Mean values over two to seven days at radon in air of homes during the heating season. Asterisks are for uncorrected for ventilation while circles are corrected for ventilation.

Figure 1

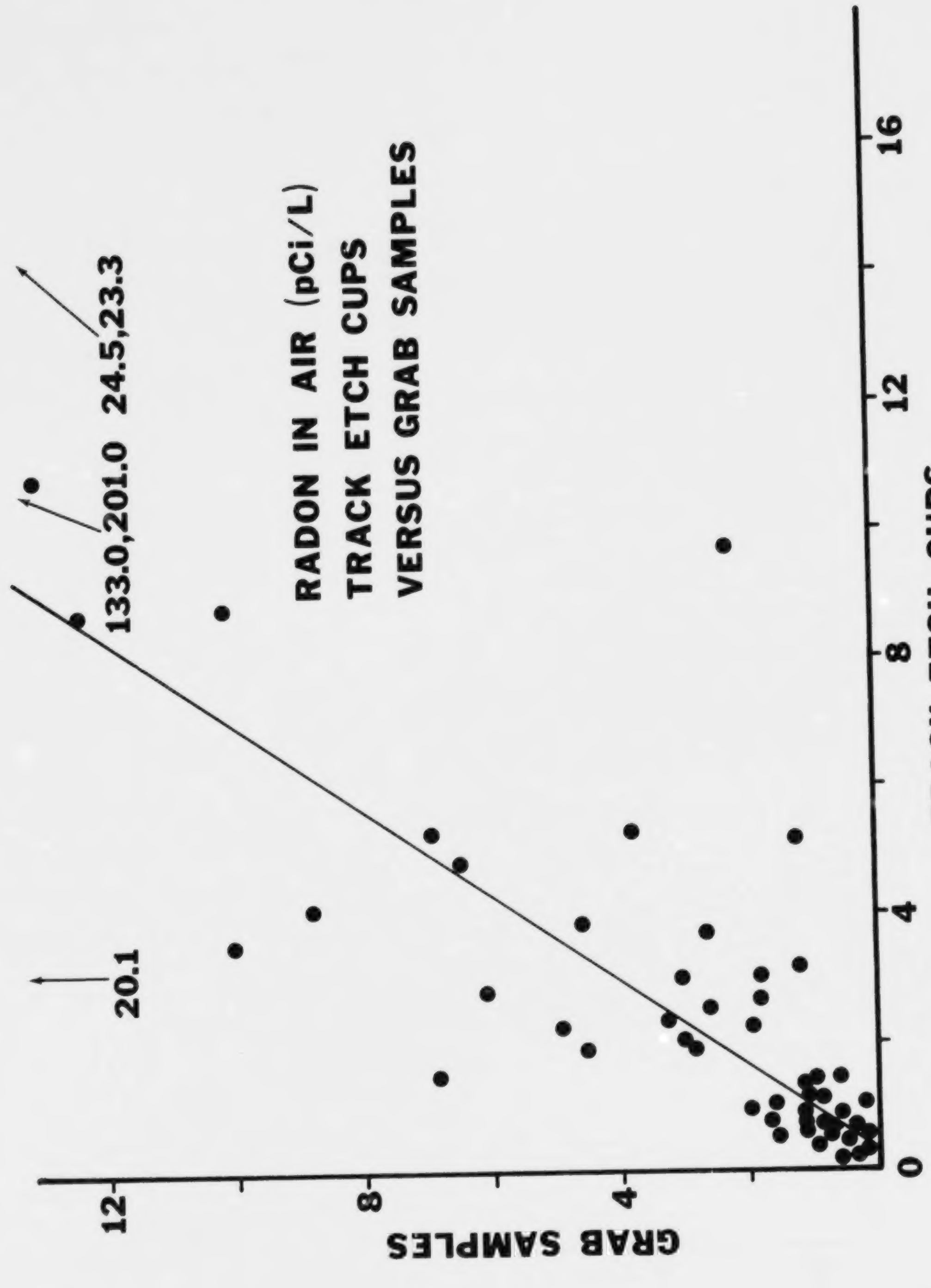
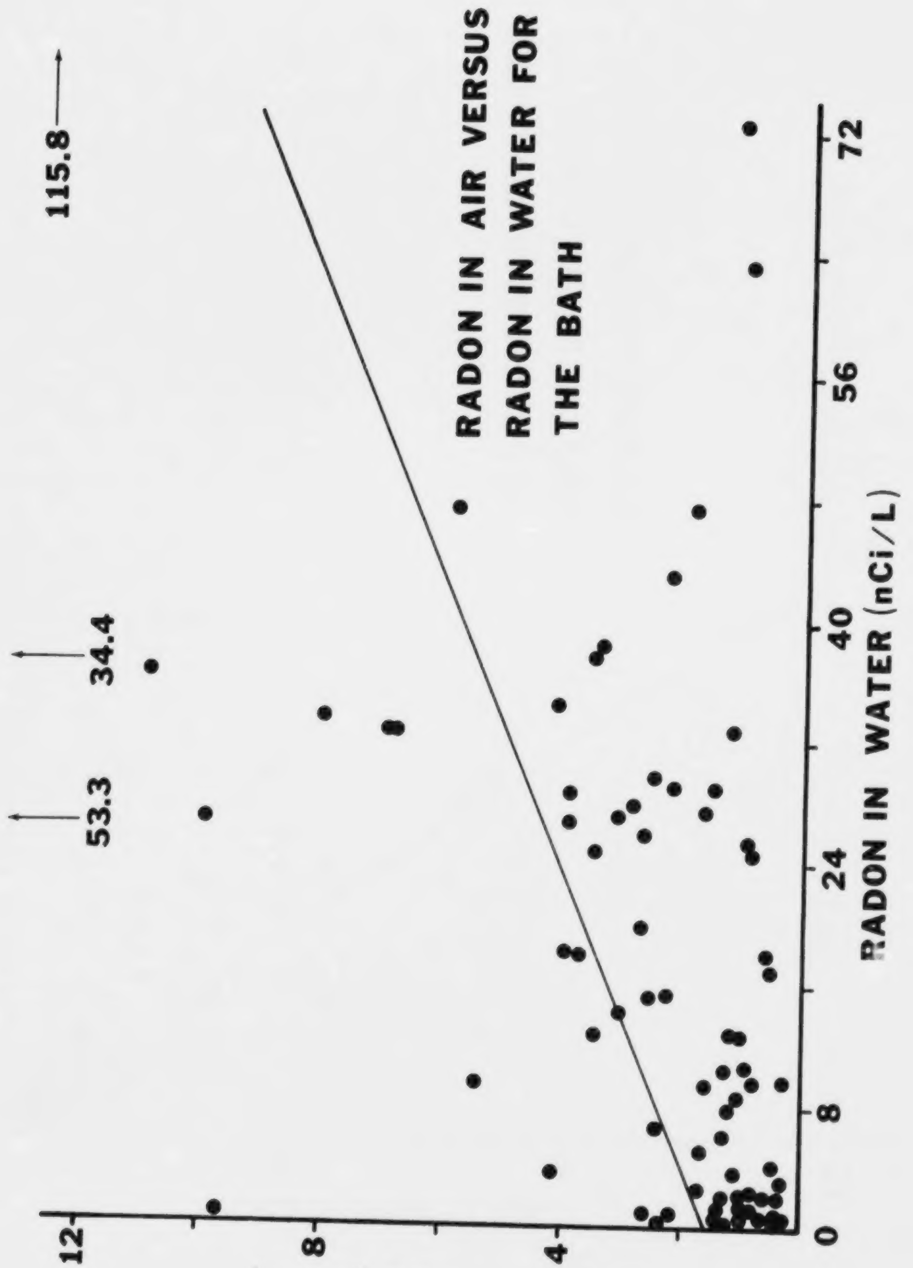
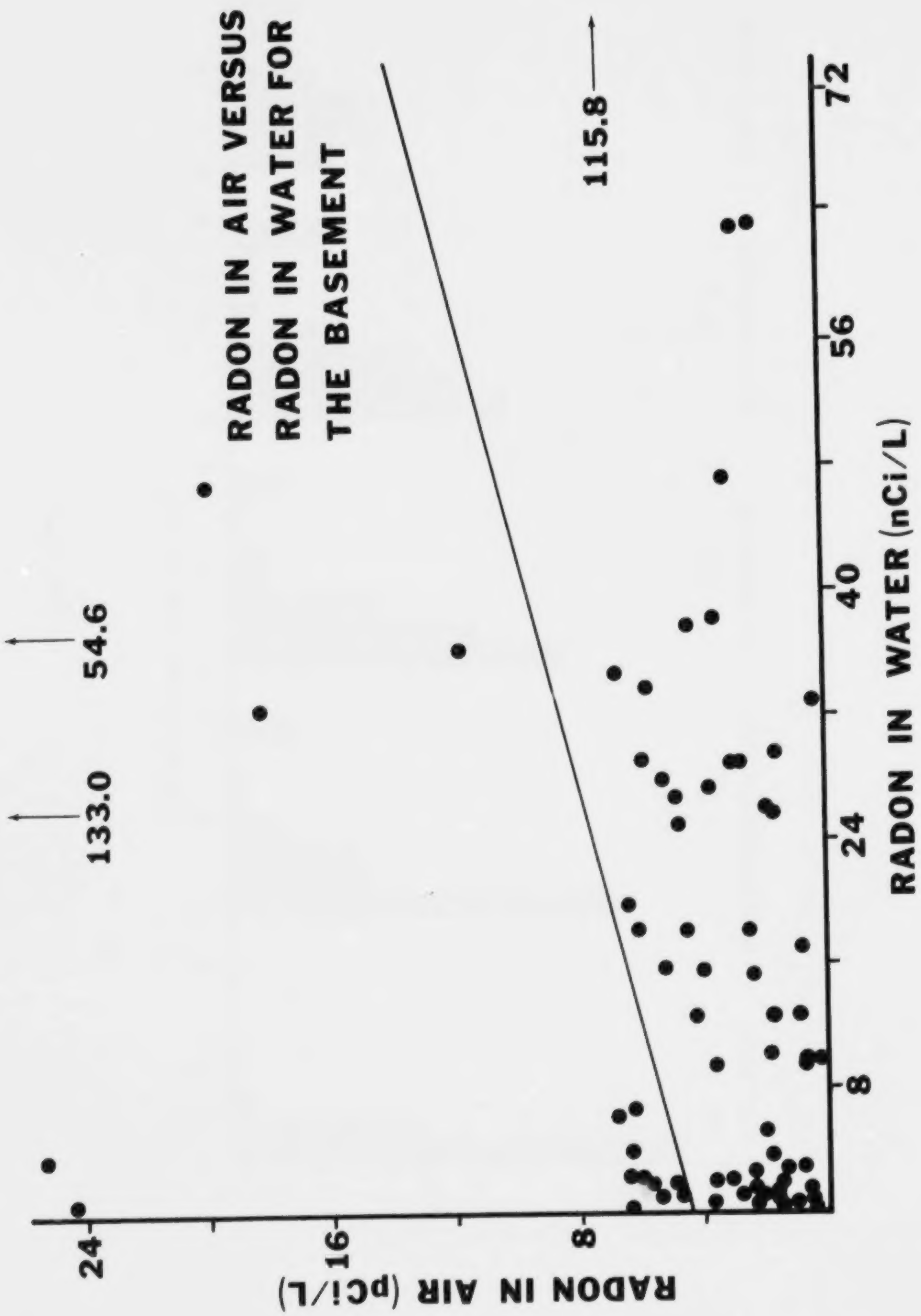

RADON IN AIR (pCi/L)
TRACK ETCH CUPS
VERSUS GRAB SAMPLES
133.0,201.0 24.5,23.3
20.1
TRACK ETCH CUPS
0
4
8
12
16
GRAB SAMPLES
4
8
12

Figure 2



RADON IN AIR VERSUS RADON IN WATER FOR THE BATH

Figure 3


RADON IN AIR VERSUS
RADON IN WATER FOR
THE BASEMENT
133.0
54.6
115.8
RADON IN AIR (pCi/L)
24
16
8
RADON IN WATER (nCi/L)
8
24
40
56
72

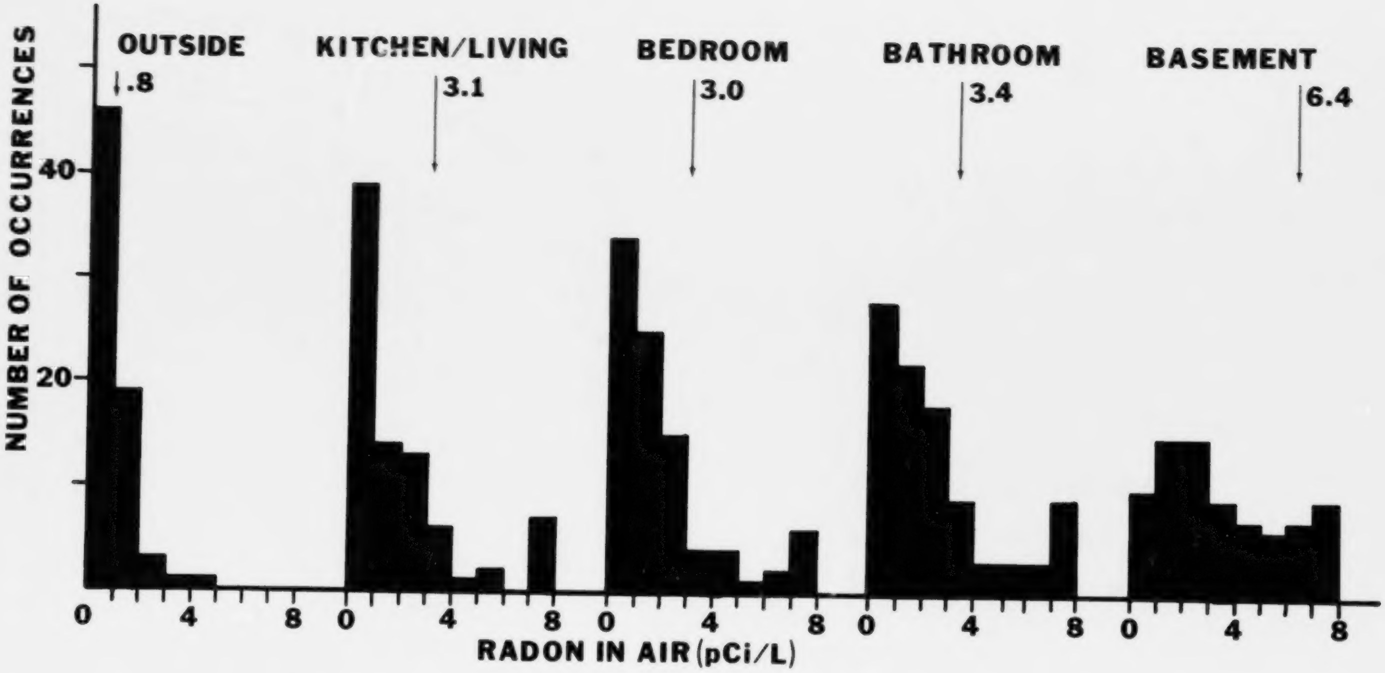

NUMBER OF OCCURRENCES
40
20
OUTSIDE
.8
KITCHEN/LIVING
3.1
BEDROOM
3.0
BATHROOM
3.4
BASEMENT
6.4
0
4
8
0
4
8
0
4
8
0
4
8
0
4
8
RADON IN AIR (pCi/L)

Figure 4

Figure 5

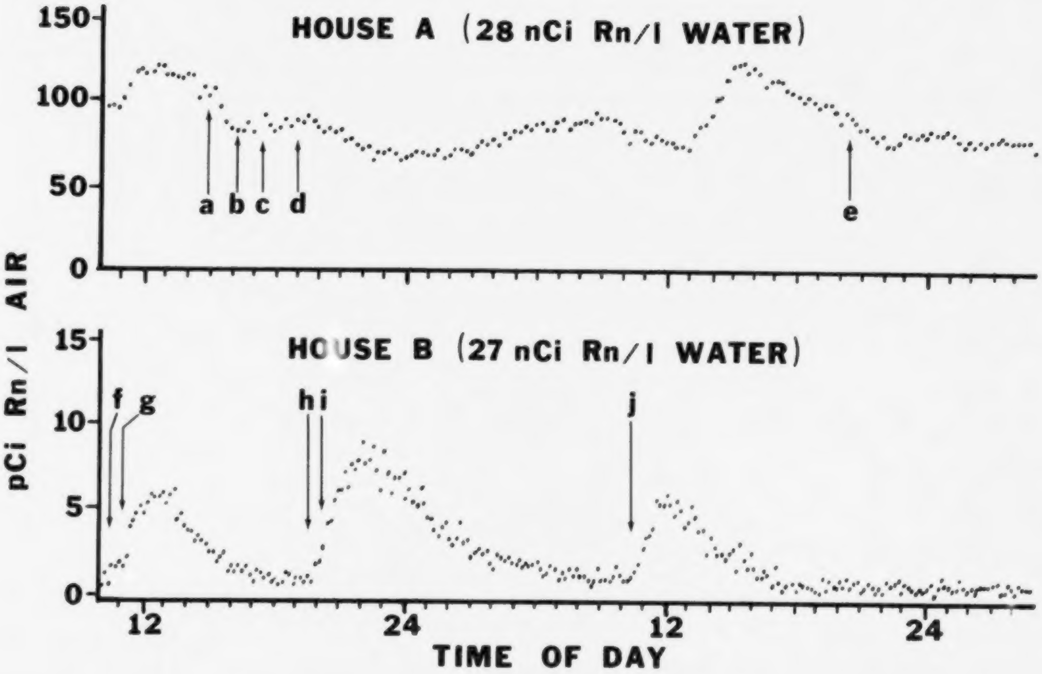
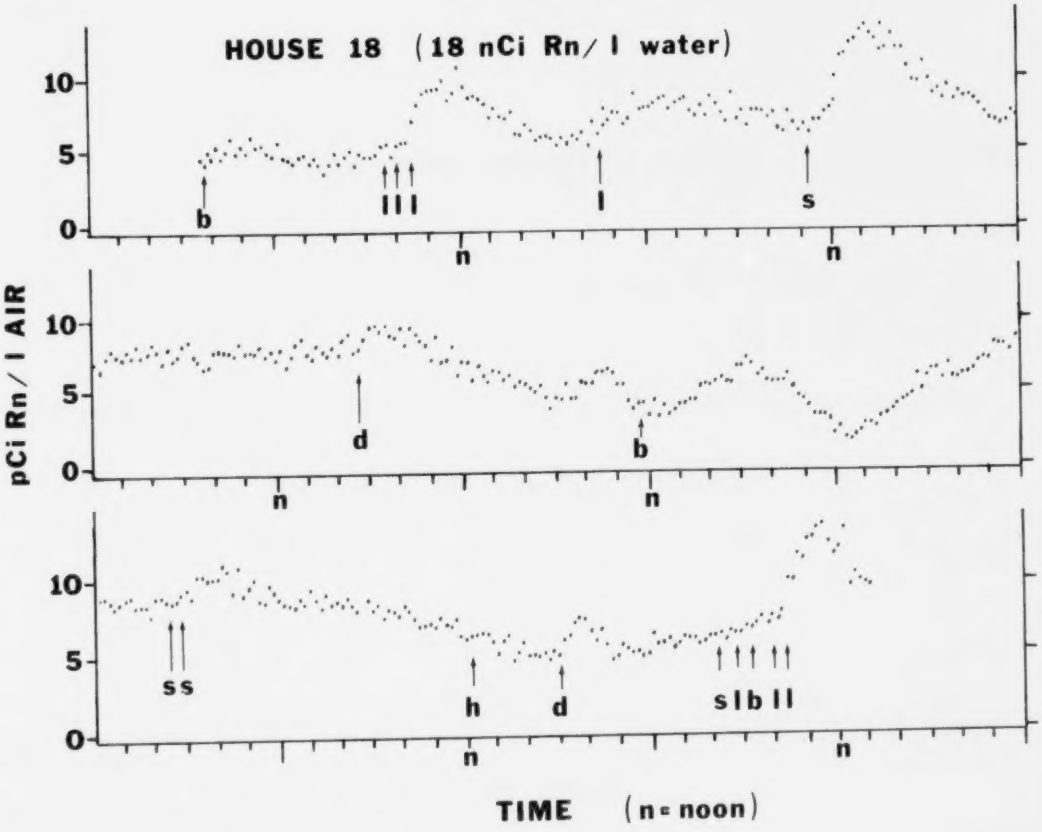
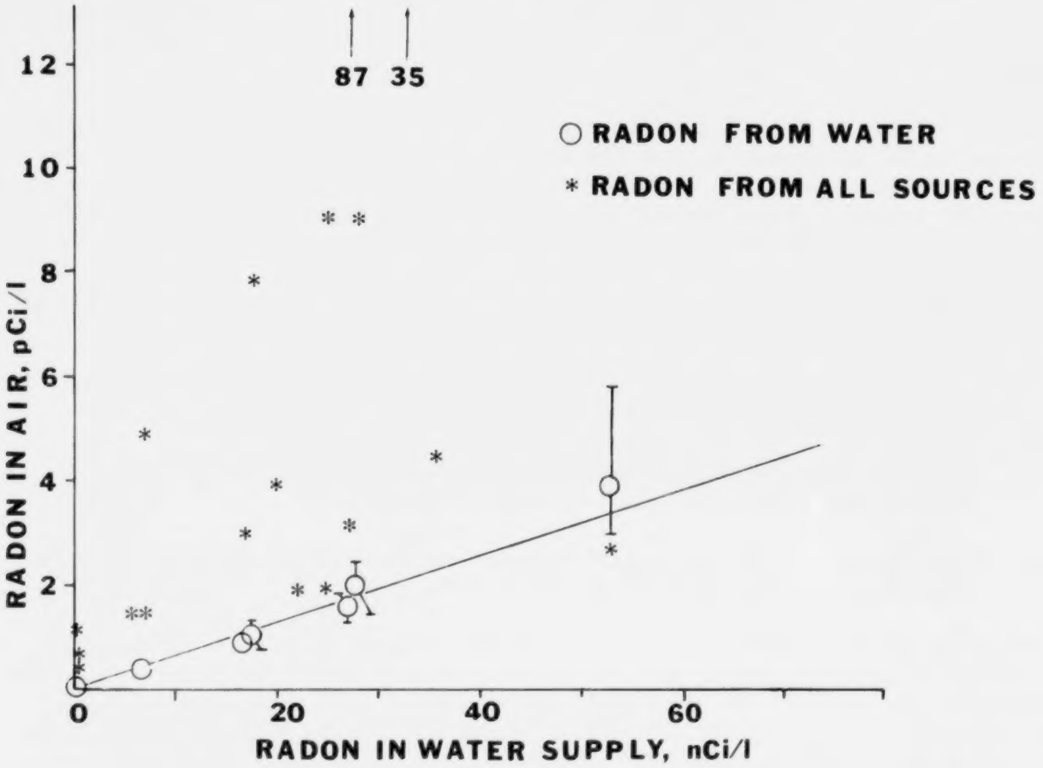

HOUSE A (28 nCi Rn/l WATER)
150
100
50
0
a
b
c
d
e
pCi Rn/l AIR
HOUSE B (27 nCi Rn/l WATER)
15
10
5
0
f
g
h
i
j
12
24
12
24
TIME OF DAY

Figure 6



HOUSE 18 (18 nCi Rn/ l water)
pCi Rn/ l AIR
10
5
0
b
l l l
l
s
n
n
d
b
n
n
s s
h
d
s l b l l
n
n
TIME (n= noon)

Figure 7


87
35
○ RADON FROM WATER
* RADON FROM ALL SOURCES
RADON IN AIR, pCi/l
12
10
8
6
4
2
0
20
40
60
RADON IN WATER SUPPLY, nCi/l

SURVEYS AND DECISION MAKING IN A REMEDIAL ACTION PROGRAM

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ABSTRACT

As the observed frequency distribution of grab WL measurements in houses was found to be lognormal with a GSD of 2.0, lognormal sampling theory was used to guide the choice of sampling frequency for an extensive survey program in over 500 houses. Analysis of lognormal data is illustrated by a Monte Carlo sampling simulation.

The tasks of the survey portion of the Remedial Action Program at Elliot Lake are:

1. identify those houses with annual average WL's in excess of the action criterion of 0.02 WL,
2. demonstrate that the annual average WL is below 0.02 WL in those houses where remedial work is not carried out,
3. demonstrate that the annual average WL following remedial work is less than the action criterion of 0.02 WL.

The program started in July 1977, and the belief at that time was that the elevated WL's in houses were due to the presence of radioactive contamination adjacent to the houses. As a result, relatively few WL measurements were made, the major effort being in gamma-ray survey. By late 1977 it was clear that radioactive contamination was not the major cause of elevated WL, and a systematic WL survey would be required to identify problem houses. The survey would have to be based on repeated grab-samples, as that was the only equipment available at the time.

By then sufficient WL measurements had been made to show that the distribution of WL measurements over a period of time in individual houses was nearly lognormal with a geometric standard deviation (GSD) of approximately 2 (see Figure 1). As a result of this, sampling theory for the lognormal distribution was reviewed to determine sampling strategy as a basis for survey organization. Fortunately many pollutants also have a lognormal distribution, and so our task was greatly simplified by the use of a NIOSH publication (NI 75) on this subject.

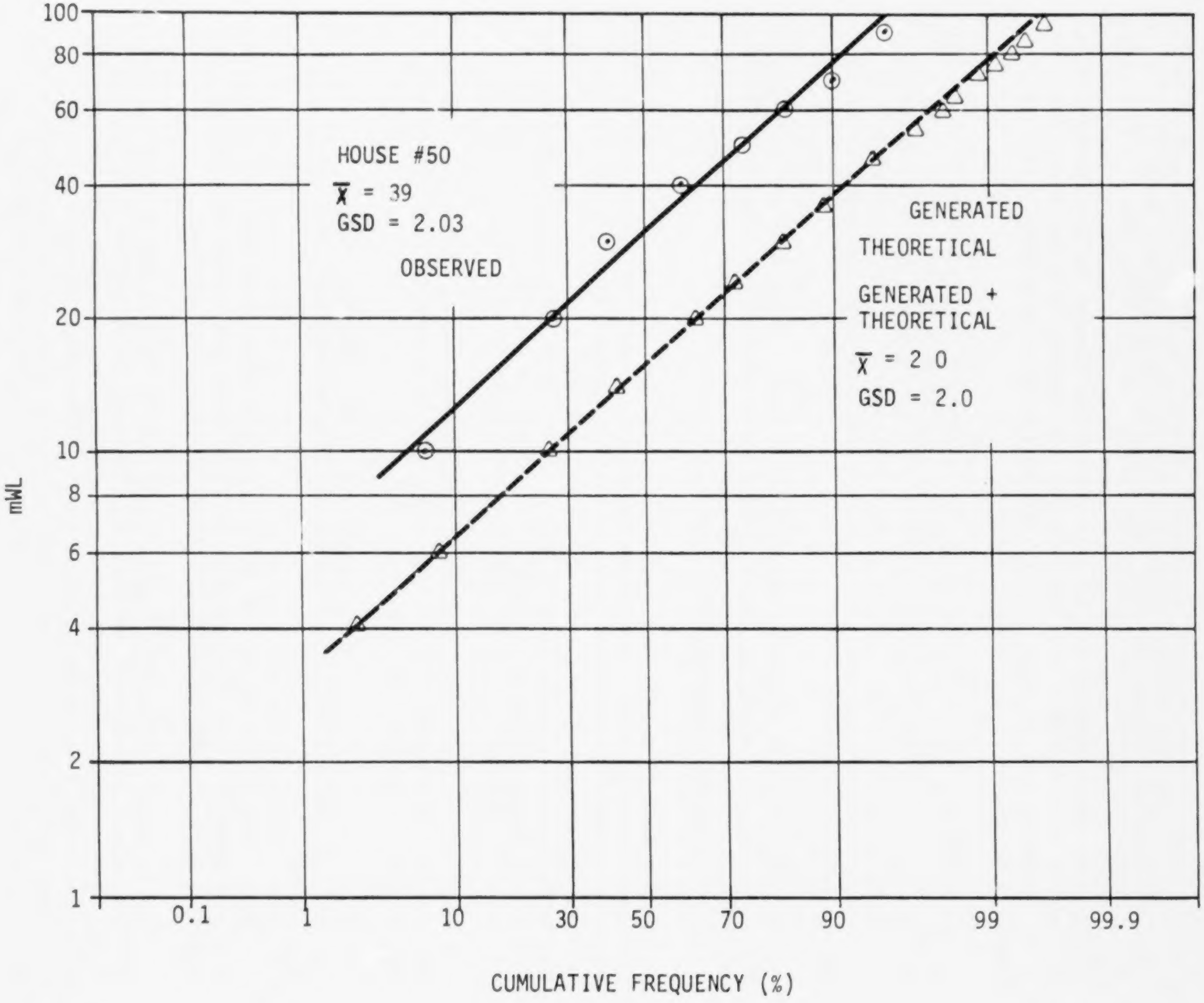
HOUSE #50
$\overline{X}$ = 39
GSD = 2.03
OBSERVED
GENERATED
THEORETICAL
GENERATED +
THEORETICAL
$\overline{X}$ = 2 0
GSD = 2.0
mWL
100
80
60
40
20
10
8
6
4
2
1
0.1
1
10
30
50
70
90
99
99.9
CUMULATIVE FREQUENCY (%)

FIGURE 1

Figure 2 is copied from (NI 75) and shows the variation of the 2 sigma confidence limits on the mean of a lognormal distribution with the number of samples taken and the GSD of the distribution. For a GSD of 2.0, at least 9 measurements are required to reduce the confidence limits to below 50%, and a large number is required to reduce the limits to less than 40%. From a practical point of view, there was little gain in accuracy in taking more than 10 independent measurements, and so that number was chosen as the minimum number of samples in a survey. For convenience, the "annual average survey" was set at 13 measurements, as this fitted with an initial measurement followed by a sample a month over a 12 month period.

As readings tend to be higher in the summer, it is necessary to take readings over a full year to avoid biasing the results according to season. There will still be a slight theoretical bias in the estimate, for the samples are not truly distributed at random, since they are not taken at night nor at weekends which together comprise nearly 3/4 of the year. However, this is unavoidable in practice.

To avoid waiting to start remedial work until a year's survey was completed, a set of rules based on "distribution free" statistics was generated to identify those houses where the annual average was clearly in excess of 20 mWL so that they could receive prompt remedial work.

When enough readings have been made to end the survey, the average WL in the house is calculated. For those houses where the average is less than or equal to 20 mWL, the data are sent to the RAP manager at the AECB* for review. If he agrees that the average is probably less than 20 mWL, he issues the homeowner a letter to that effect. If he feels that the calculated average is not fully representative of the average in the house, he may request extra survey readings, or that remedial action be carried out in the house.

For those houses where the average is greater than 20 mWL, the data are sent to the RAP manager for review. The AECB then contact the homeowners to obtain their agreement to remedial work.

After the remedial work is completed, a Post Remedial Survey is carried out to show that the WL in the house has been successfully reduced to less than 20 mWL.

Major restoration of finish and appearance is not started until this survey is completed. As the homeowner is naturally interested in completion of remedial work as soon as possible, a rapid answer is required.

The procedure adopted is to take 1 WL measurement a day for 10 consecutive working days. If the best estimate of the mean is less than 0.02 WL, the house is forwarded to the RAP manager as probably complying with the primary criterion. The Compliance Division of the AECB subsequently check these houses to verify that the WL has been reduced. The interrelationship between the survey, the remedial program, and the AECB is shown in Figure 3.

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* AECB-Atomic Energy Control Board. The AECB directs the Remedial Action Programs carried out and financed by the Joint Federal Provincial Task Force on Radioactivity.

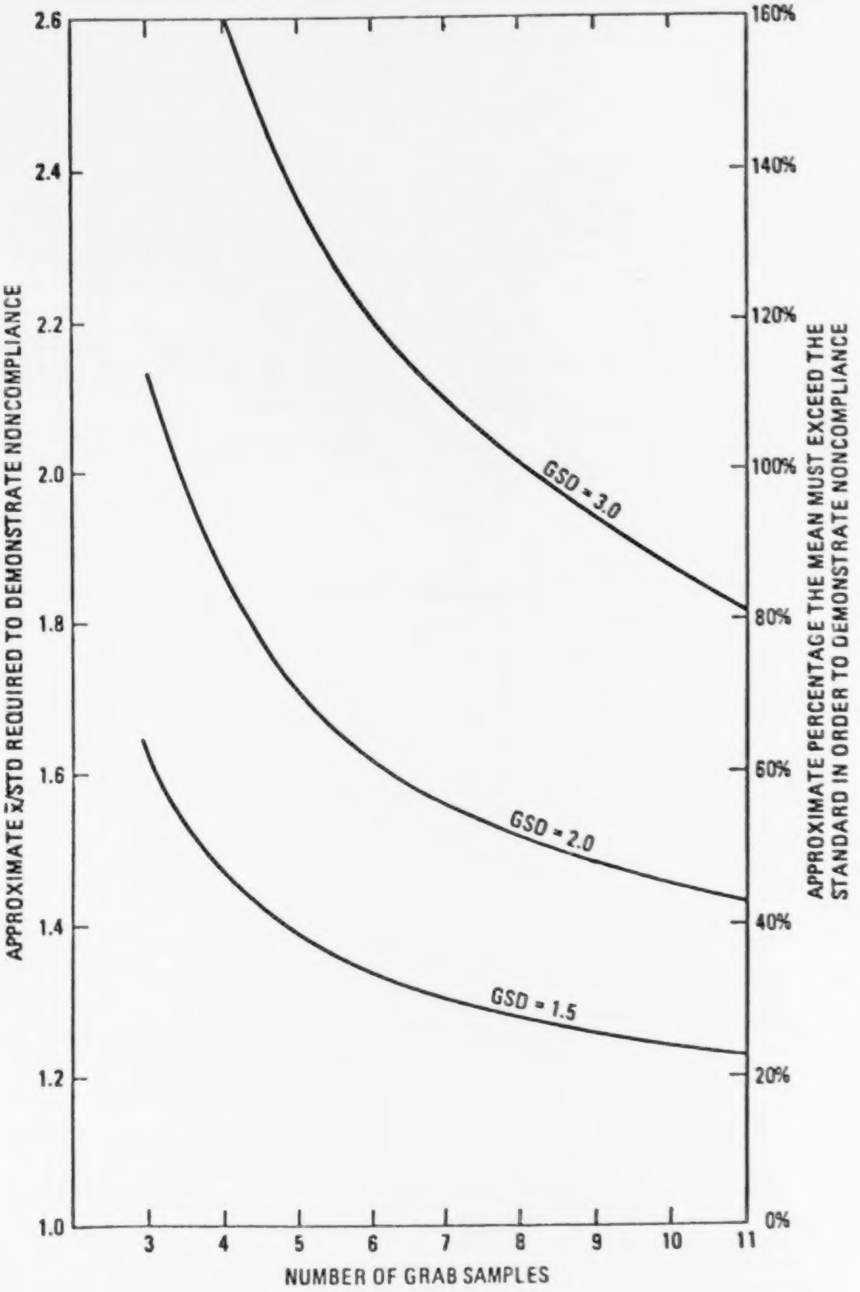


Figure 2 - Effect of small grab sample sizes on requirements for demonstration of noncompliance. Three different data geometric standard deviations (GSD) are shown which reflect the amount of variability in the environment.

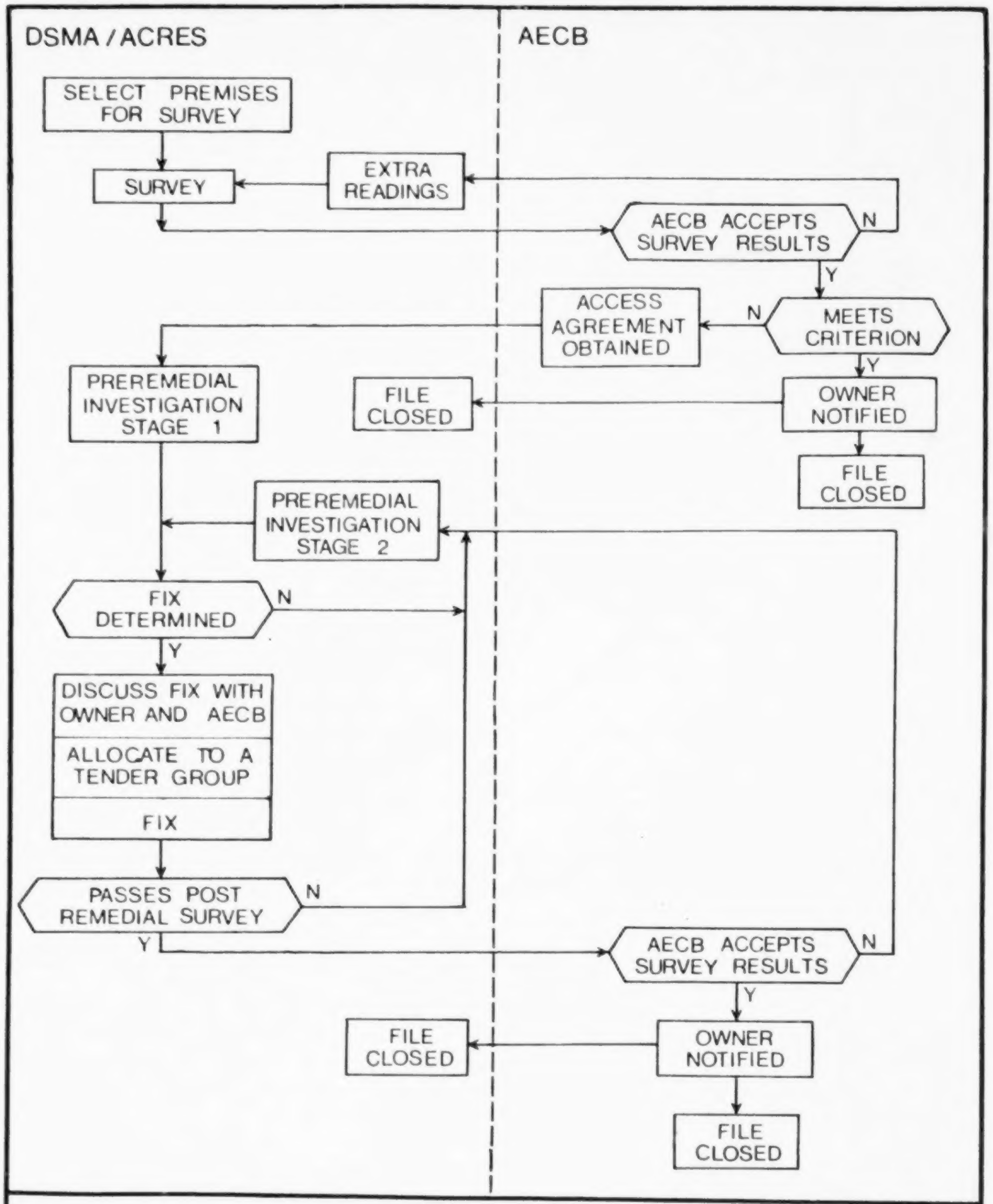

DSMA / ACRES
AECB
SELECT PREMISES FOR SURVEY
SURVEY
EXTRA READINGS
AECB ACCEPTS SURVEY RESULTS
N
Y
ACCESS AGREEMENT OBTAINED
N
MEETS CRITERION
Y
PREREMEDIAL INVESTIGATION STAGE 1
FILE CLOSED
OWNER NOTIFIED
FILE CLOSED
PREREMEDIAL INVESTIGATION STAGE 2
FIX DETERMINED
N
Y
DISCUSS FIX WITH OWNER AND AECB
ALLOCATE TO A TENDER GROUP
FIX
PASSES POST REMEDIAL SURVEY
N
Y
AECB ACCEPTS SURVEY RESULTS
N
Y
FILE CLOSED
OWNER NOTIFIED
FILE CLOSED

FIGURE 3

REMEDIAL PROGRAM LOGIC DIAGRAM

As a large number of houses (500+) were under survey, it was not practical to keep an overview of the program by manual summaries. Therefore, a computer accessible data base was developed which lists the survey readings made in a house, produces estimates of the house average WL and also keeps track of the progress of the house through the remedial action program.

A demonstration of the analysis problems posed by the lognormal distribution, a Monte Carlo sampling simulation was carried out. Figure 4 shows a histogram of 5500 numbers generated randomly from a lognormal distribution with mean 20 and GSD of 2.0. If these 5500 lognormal numbers are analyzed in the usual manner (which assumes a gaussian distribution) the mean is 19.7 and the standard deviation is 15.6. The gaussian with these parameters is shown in Figure 4, and can be seen to be a rather inadequate description of the actual distribution of numbers. Approximately 16% of the numbers would have to be negative for the gaussian to apply.

However, we do not make decisions based on a single reading, but only on the average of groups of readings. It is our practice to take at least 10 readings in a house before we make any statistical decision, and so it is the frequency distribution of samples of 10 that is of interest. This distribution was obtained by taking 550 groups of 10 numbers from the 5500 lognormal numbers generated earlier. The arithmetic mean and the best-estimate mean were calculated for each group of 10. The best-estimate mean is based on the logarithmic mean of the sample numbers, and is the theoretically correct method to estimate the average value of a lognormal distribution from a sample. Figure 5 shows the CDF of the 550 best-estimate means. They are lognormally distributed as expected, with variability reduced close to the theoretical value of GSD = 1.25. The distribution of arithmetic means was virtually identical to this, showing that the uncertainty in the estimate of the mean was not a function of the method of analysis, but of the variability of the measurements themselves. The slight asymmetry in the distribution does require the confidence limits to be asymmetric, and so confidence limits based on gaussian analysis will be incorrect. For example, gaussian analysis of the 5500 numbers gives 95% confidence limits on the mean groups of 10 samples as 11.6 and 27.8. Figure 5 enables the 95% confidence limits to be read directly. They are approximately 13 and 28.5, which are close to the gaussian values, but slightly higher.

In practice the use of confidence limits creates yet another problem - what to do in the case of those measurements that fall between the confidence limits. For example, if we wish to work to 90% two tailed confidence limits, and we make 10 measurements, Figure 5 shows us that we can make no decision if the mean of the measurements is more than 13 mWL and less than 28.5 mWL. Now, the one thing we cannot do in the real world is to take measurements and then fail to make a decision, and although we can narrow the limits by taking more readings, the zone of indecision cannot be removed completely except by an arbitrary decision. We prefer to make our arbitrary decision first, and so for our surveys we work at 50% confidence. At this level there is no zone of indecision, and it has the intuitively appealing feature that we apparently believe the average of our measurements when it comes to deciding if the annual average WL in a house is, or is not, greater than 20 mWL.

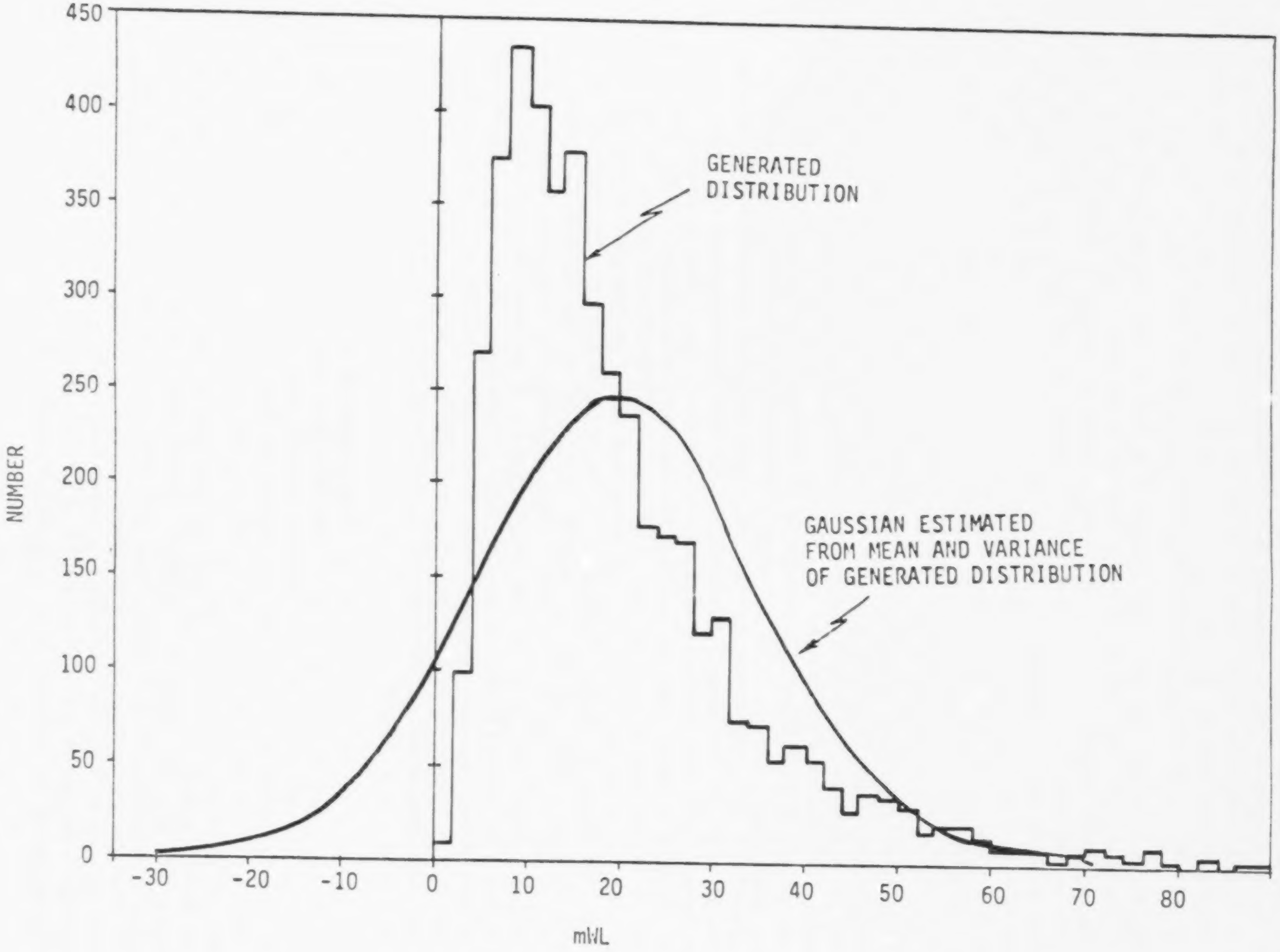
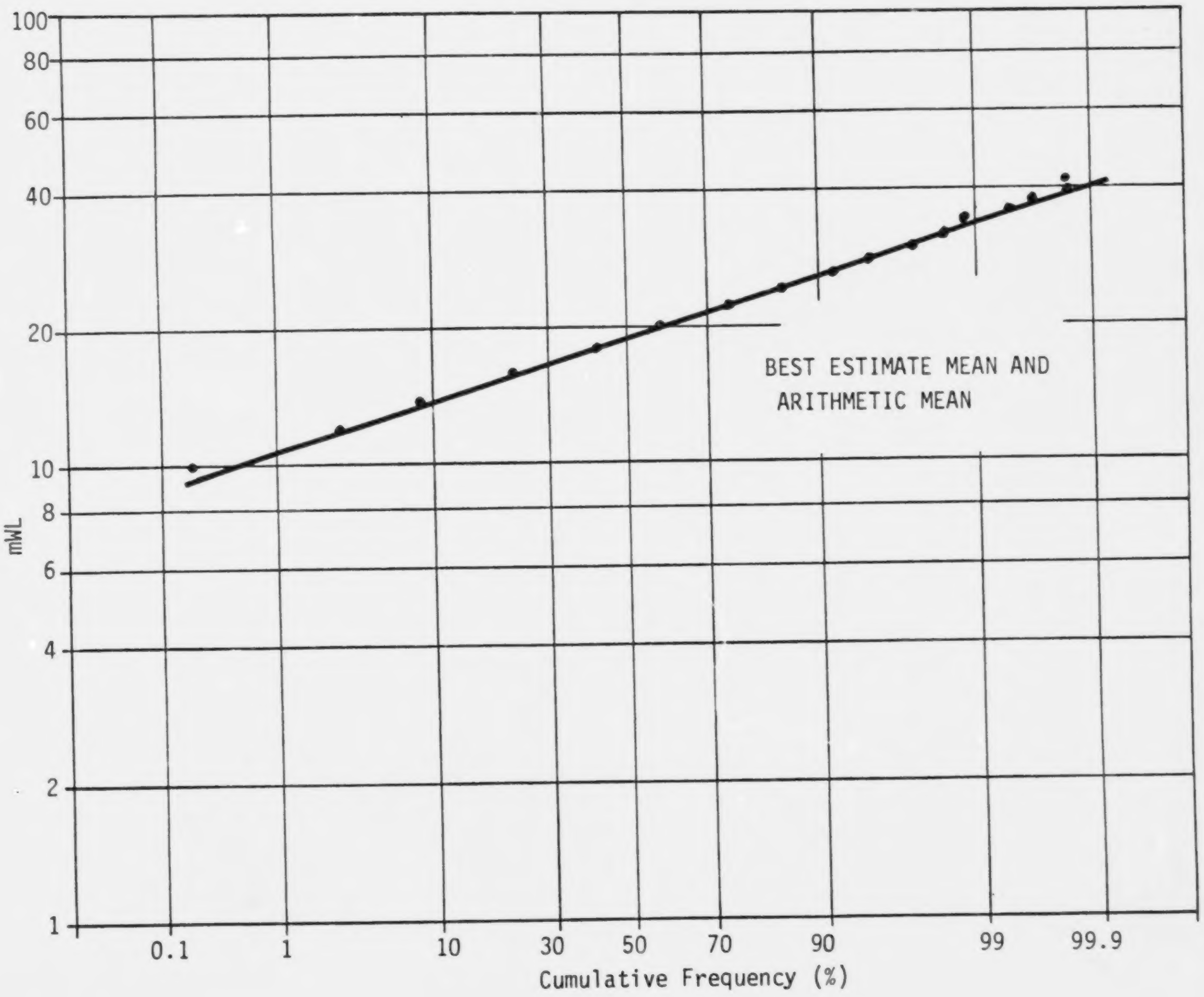

GENERATED
DISTRIBUTION
GAUSSIAN ESTIMATED
FROM MEAN AND VARIANCE
OF GENERATED DISTRIBUTION
NUMBER
450
400
350
300
250
200
150
100
50
0
-30
-20
-10
0
10
20
30
40
50
60
70
80
mWL

FIGURE 4



DISTRIBUTION OF MEANS - GROUPS OF 10

FIGURE 5

REFERENCES

Ni 75 "Statistical Methods for the Determination of Non-compliance with Occupational Health Standards." HEW Publication No. (NIOSH) 75-159, April, 1975.

AIRBORNE Rn-222 IN BUILDINGS CONSTRUCTED WITH HIGH-RADIUM-CONTENT CONCRETE BLOCKS

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ABSTRACT

The relation of Rn-222 in building air to Ra-226 in structural materials was examined by measuring the Rn-222 flux density at surfaces and concentration in air in two office and laboratory buildings. The buildings have walls constructed partially with concrete blocks that contain Ra-226 at relatively high levels. The Rn-222 flux density at walls and floors was measured by gamma-ray spectral analysis of charcoal collectors for radon that had been sealed to surfaces for 3-day periods. The Rn-222 concentration in air was measured by collecting air in evacuated 125-cc radon scintillation (Lucas) cells and counting alpha particles in the cells. Results were compared with samples collected outside and in a building that did not have elevated Ra-226 levels in structural materials. To determine source terms, Ra-226 concentrations were measured in building materials and soil. Contributions to the Rn-222 concentration in building air from the ground beneath the building, outside air, and construction materials were estimated. The identified sources contributed between 67 and 100 percent of the average of measured concentrations; in every instance, most of the Rn-222 was from the ground beneath the building.

INTRODUCTION

Some concrete made from phosphate slag contains elevated Ra-226 levels. Buildings constructed with this concrete were identified in surveys through increased external radiation exposure rates due to the gamma radiations that Ra-226 and its Pb-214 and Bi-214 progeny emit. Exhalation of the gaseous Rn-222 daughter from concrete walls, floors, or pillars into building air will also increase the radiation dose to the lungs. This dose is due to alpha particles emitted by the Po-218 and Po-214 progeny, at concentrations usually specified in terms of the working level (WL). An increased WL value is generally considered to indicate a greater hazard than the accompanying external exposure rate in a building, but is not as easily quantified because radon and its progeny in building air fluctuate widely. Complicating factors in determining Rn-222 levels in air due to building materials are the dependence of these levels on the ventilation rate and the fraction of Rn-222 emanating from the material, as well as relatively high levels from other sources. This report describes measurements of Rn-222 concentrations in air and exhalation rates from surfaces to distinguish among the sources of Rn-222 in air in three buildings, two of which were built with concrete blocks that contain high-radium phosphate slag.

The slag from the thermal process for phosphorus production is a calcium silicate that has been used variously for the fine and coarse aggregate in concrete (Bo77, He79). Phosphorus plants in Idaho (Bo77, Pe78), Montana (Ll78) and Alabama (Ma78, Ka79) have produced slags that contain Ra-226 at concentrations between 30 and 60 picocurie per gram (pCi/g). The Ra-226 concentration in concrete depends on the Ra-226 level in the slag derived from phosphate rock and on the fraction of slag used in the concrete. Radiation exposure rates of approximately 40 microroentgen per hour (uR/hr) have been measured in structures built with this material (Bo77, Ka79). In comparison, exposure rates in buildings due to cosmic rays and Ra-226, Th-232, and K-40 in building material and the ground usually range from 6 to 18 uR/hr (Ka79).

The EPA had proposed rules under the Resources Conservation and Recovery Act of 1976 to prevent use of byproduct materials such as phosphate slag if they increase gamma ray exposure rates by 5 uR/hr or WL values by 0.03. At equilibrium, 0.03 WL results from a Rn-222 concentration of 3 picocurie per liter (pCi/L). In buildings, the ratio of WL to Rn-222 concentrations normally is well below this value, but differs according to various conditions. The short-lived progeny do not attain equilibrium due to air turnover and deposit on surfaces to some extent. Mixtures of Rn-222 and progeny in building air typically result in approximately 0.004 WL per pCi/L (UN77). Increases of 5 uR/hr and 0.03 WL or the corresponding 7.5 pCi/l are readily detected.

A number of reviews have considered Rn-222 in building air and its sources (Co81, Ea75, Ha78, Ka79, Le81, Mo76, NC75, OE79, Ta80, UN77). Elevated Rn-222 concentrations in building air can be due to high concentrations in outside air, the ground beneath, or the water or gas supply. Initially low levels can rise steeply due to accumulation of radon and its daughters within a tightly closed room or structure (Sp80). The Rn-222 concentration in outside air is typically a few tenths of a pCi/L, but may fluctuate in the course of the year by almost two orders of magnitude at a single location (Ha78a). Radon inflow from beneath the building is particularly apparent in houses built on uranium tailings and at locations of former phosphate mines (Gu80, Ea75).

Average WL values in buildings not known to be associated with any elevated Ra-226 levels appear to be between 0.003 and 0.007 WL (Ka79, Ge80). Because of the above-cited factors, the individual values on which these averages are based may vary by almost two orders of magnitude among the buildings included in a particular survey.

Radon-222 progeny levels in buildings constructed with high-radium concrete were between 0.0005 and 0.05 WL in 107 structures in Idaho (Pe78); between 0.001 and 0.1 WL (except for one higher value) in 69 Montana houses (Ll78); and averaged 0.018 WL in 17 Alabama houses (US80). These measurements were performed for extended periods to determine average radiation doses. In Montana, averages in two towns were 0.02 and 0.013 WL (US80). The log-mean value of 0.006 WL in the Idaho town is reported to be the same as the normal background, and the corresponding log-mean Rn-222 concentration of 1.4 pCi/L is also approximately the same as in normal structures (Pe78, US80). The Rn-222 concentration and WL value in outside air and the Rn-222 flux density from the ground beneath buildings were not reported.

In this study, air samples for Rn-222 analysis were collected in brief (1-minute) periods at 3 multi-story office/laboratory buildings located near each other. In building NE, interior walls on the second and third, but not the first floor, are of concrete blocks with a Ra-226 concentration of 19 pCi/g; in building ER, most walls, interior and exterior, on all three floors were constructed with these blocks; while in building CE the building materials are not elevated in Ra-226. The Ra-226 in all concrete except the blocks was estimated to average 1.5 pCi/g on the basis of external radiation exposure rates (Ka79). Air samples were collected simultaneously to compare Rn-222 levels inside and outside, on the various floors of individual buildings, and on corresponding floors in the three buildings. Samples were collected in the mornings, when the highest levels for daytime occupancy were expected due to the early-morning peak in Rn-222 concentrations outside and the overnight accumulation inside while the buildings were closed. The turnover rate of air within the three buildings was measured by releasing and collecting a tracer gas.

The exhalation rate of Rn-222 from the concrete blocks was measured for 3-day periods in the NE and ER buildings and compared with that from poured concrete walls in the NE and CE buildings. The Ra-226 content and Rn-222 emanation fraction (or emanation power) in crushed blocks was also measured. The exhalation rates of Rn-222 from soil and through the floors of the three buildings were measured to determine the relative contribution by the ground to the total Rn-222 concentrations in air. The external radiation exposure rate and Ra-226 concentration in surface soil were measured as possible indicators of the magnitude of Rn-222 exhalation rates from the ground. The Rn-222 concentration was measured in the public water supply to determine the contribution of this potential source to radon levels in building air.

PROCEDURE

The 3 measurement sites are combined office and laboratory buildings at the Georgia Institute of Technology. The NE building is built on a slab and is three stories high. Its outside walls are brick with some concrete and metal panels; the inside walls are mostly of concrete block. The second and third stories were added in 1965. Only the blocks on these floors contain elevated amounts of Ra-226. The ER building was also built in 1965, except for a small west wing added later. It has a basement that is above ground on two sides, 2 floors above basement level, and a small partial third floor. The exterior walls are concrete block faced with brick and some concrete and metal panels above the basement; the interior walls are of concrete block. Except in the newer wing, the blocks have the same elevated Ra-226 levels as those in the NE building. The CE building, used as control, is much older. It has a basement that is above ground on two sides, and three stories above the basement. The walls are of poured concrete, faced with brick. All buildings have structural supports and floors of poured concrete. The two newer buildings are centrally air conditioned. The older building has window or area air conditioners in most offices and laboratories, but the hallways and a laboratory extending throughout the basement are not air conditioned.

Cylindrical cells (6.0-cm dia. x 9.4 cm outside) coated on the inside with ZnS(Ag) scintillator were used to measure alpha particles from radon and its progeny (Gu80). The cells have a quartz window at one end and a glass stopcock at the other. The cells were evacuated to approximately 0.04 atmosphere with a vacuum pump before sample collection. They were counted within a shield to keep out light by placing the window on a photomultiplier tube. The cells were calibrated with standard Ra-226 solutions at 5 pCi/L levels. The counting efficiency for combined Rn-222, Po-218, and Po-214 alpha particles was 2.5 ± 0.1 counts/disintegration of Rn-222. Before and after each measurement of Rn-222, the detector background was determined. Each sample was counted twice in succession. The presence of Rn-222 and the absence of interference was confirmed by observing the 3.82-day half life of Rn-222. The combined counting periods of the background measured before and after counters were typically 1,200 minutes, and the two sample counts were for a similar period. Intervals of 150 minutes between sample collection and measurement and between cell flushing and background measurement were maintained to assure decay of any unsupported short-lived radon progeny.

Air samples were collected by opening the stopcock of an evacuated cell for approximately 1 minute. The cell was usually held 1 m above the floor in the middle of corridors. Outdoors, air was collected 1 m above open ground and several meters distant from the building. Weather conditions were recorded.

The standard deviation of counting these samples, which is believed to be the major source of random error, was typically 0.04 pCi/L, and the minimum detectable level, defined as the 3-sigma counting error of the background, was 0.08 pCi/L. Three samples collected side by side on 9-6-81 (see Table 1) measured 0.85, 0.81, and 0.86 pCi/L. Hence, differences by more than 0.1 pCi/L are considered significant.

Table 1

Radon-222 Concentrations in Air, pCi/L

			CE Building					NE Building				ER Building			
Date, 1981	Time	Conditions	out-side	base-ment	1st	2nd	3rd	out-side	1st	2nd	3rd	out-side	base-ment	1st	2nd
3-6	0910	S, NC, BW*						<0.1 <0.1	1.0 1.0	0.6 0.5					
3-12	0845	S, NC, BW	<0.1	0.8 0.3		1.0									
3-17	0900	S, NC, LW						0.2	1.5	0.9 1.0	0.6				
3-19	0900	S, NC, BW										0.1	0.5 0.4	0.1 1.1	0.9
3-23	0820	OC, LW, rained earlier	0.1	0.7	0.4	0.5									
7-8	1040	S, NC, LW	<0.1	0.4 0.9		0.1	<0.1								
7-10	1040	S, NC, C						0.2	1.1 1.5	0.6 0.7					
7-15	1030	S, NC, LW													
												<0.1	3.2 3.7	2.9	2.2
7-21	1050	S, NC, LW	0.1	0.2					1.3			0.2	2.9		

Table 1 (cont'd)

Date, 1981	Time	Conditions	CE Building					NE Building				ER Building			
			out-side	base-ment	1st	2nd	3rd	out-side	1st	2nd	3rd	out-side	base-ment	1st	2nd
7-25	1040	S, NC, LW	<0.1	0.6				0.3	1.8			0.2	3.5		
7-28	0730	S, NC, LW						0.4	1.6						
	0930							0.2	1.3						
	1130							<0.1	0.9						
7-30	0730	OC, LW						0.2		0.8					
	0930	light rain						0.3		0.5					
	1130	at 11:30								0.6					
8-4	0810	S, NC, C	0.3	1.9	0.5	0.4	0.6								
8-6	0750	S, SC, LW						0.2	1.3	0.9	0.8† 0.8 0.9				
8-13	0830	S, SC, LW										0.3	2.6	2.9 3.2	2.1 2.2

* S: sunny; NC: no clouds; SC: scattered clouds; OC: overcast; C: calm; LW: light wind; BW: brisk wind;
† triplicate samples

The Rn-222 content in tap water collected in the CE building on 11-17-81 was measured in duplicate samples. Aliquots of 18 ml water were placed in emanation tubes and radon was flushed from them into Lucas cells, which were counted twice in succession for a total period of 1,300 minutes. The background in these cells was measured before and after the samples for 500 minutes each. The lower limit of detection (3-sigma value of the background count) was 0.6 pCi/L. The same water--Atlanta city water obtained from the Chattahoochee River--is used in all three buildings.

The rate of air turnover was determined by releasing SF_6 gas on separate occasions on the lowest and second lowest floor of each of the three buildings. Approximately 1 cc of the gas was released. Air samples of 1 cc each were then collected in 2-cc plastic graduated syringes and injected into a gas chromatograph (Varian model 3700). The SF_6 peak was observed just before the oxygen peak. The peak areas were measured for samples collected generally at 10-minute intervals for one to two hours after gas release. These areas were plotted on semilogarithmic paper as a function of time after gas release to determine the air exchange (ventilation) rate, λ_a, in hr^{-1}.

The Rn-222 flux density was determined by collecting radon on activated cocoanut charcoal (6 - 14 mesh) in cylindrical plastic containers and counting the charcoal after ingrowth of the Pb-214 and Bi-214 progeny with a Ge(Li) detector plus multichannel analyzer. The container has a 500-cc volume and is filled with 200 cc charcoal (122 g) retained by a paper filter and metal mesh at the bottom of the container. A plastic cover seals the container before and after collection. The container was pressed tightly against the surface to be monitored and sealed to it with putty; for soil measurements, the container was pressed into the soil. Samples were collected for 3-day periods and counted for 10,000 to 50,000 seconds, depending on the count rate due to Rn-222 progeny. The count rate was corrected for background radiation due to Rn-222 progeny in air and decay of Rn-222 during collection, between collection and counting, and during counting. The measurement was converted to units of picocurie per square meter per hour (pCi/m^2hr) on the basis of the counting efficiency of the detector for a 200-cc sample volume, the period of collection, and the 80-cm^2 surface area of the collector.

Measurements of Rn-222 flux density were performed on walls constructed of high-radium concrete blocks and of poured concrete, on poured concrete floors, and on soil near the buildings. Two of the samplers were left open to air in the CE building and two were sealed with the cover for 3-day periods to test, respectively, the extent to which still air collects on the charcoal and the extent of leakage into the container. The open charcoal sampler collected the equivalent of 500 pCi/m^2hr. The closed container collected no Rn-222, at the detection limit of 10 pCi/m^2hr.

Concrete blocks from the NE, ER, and EA buildings (the latter a warehouse constructed with high-Ra-226 blocks) were broken into small pieces, ranging from powder to approximately 5 mm in diameter, and weighed 500-cc aliquots were analyzed for Ra-226 concentration by gamma-ray spectrometer. To determine the emanation fraction, a 10-gm aliquot was placed in a 50-cc glass tube with stopcocks at each end, connected to a second 50-cc tube. The sample was stored for 2 to 6 days at accumulate Rn-222 in both tubes. The connection between the two tubes was then closed and the air in the second tube (containing no concrete) was flushed into an evacuated radon scintillation flask. The measured volume was 40 percent of the air over the concrete sample. The ratio of the Rn-222 emanation rate to the generation rate was determined from the Rn-222 in the sample and the Ra-226 in the material, corrected for decay of Rn-222 during collection, between collection and measurement, and during measurement.

Soil samples were collected from open ground at the two locations where the Rn-222 flux density had been measured outside each building. Volumes of 500 cc were scraped from the top 2 cm of ground. The samples were placed in plastic 500 cc containers, weighed, and measured by gamma-ray spectrometry.

Radium-226 was determined in soil and pieces of concrete block in closed plastic containers by measuring the 352- and 609-keV gamma rays of Pb-214 and Bi-214, respectively. To check these values, the 186-keV gamma ray of Ra-226 and U-235 were also measured. It was assumed that 58 percent was due to Ra-226 and the rest due to U-235, and that the Ra-226 gamma ray represented 3.5 percent of Ra-226 decays. The 583- and 911-keV gamma rays of Ra-228 progeny were used to determine Ra-228, and the 1,461-keV gamma ray was used for determining K-40. The radium results are uncertain to approximately 5 percent because of uncertainties in the applied gamma-ray fractions. In addition, the standard deviation of the count rate--5 to 10 percent--depends on radionuclide concentrations and counting periods.

A Reuter-Stokes pressurized ionization chamber held 1 m above ground was used to measure external radiation exposure rates at the same locations. The detector was calibrated with a 10 mg Ra-226 standardized source.

RESULTS AND DISCUSSION

The measured Rn-222 concentrations in air listed in Table 1 average 0.6, 1.0 and 2.2 pCi/L respectively in the CE, NE, and ER buildings compared to 0.2 pCi/L outside. Each of these averages represents between 16 and 25 measurements. The averages are consistent with previous observations of much higher Rn-222 concentrations that have been encountered inside buildings than outside. Outside levels were near 0.2 pCi/L during extensive surveys in Sweden (Hu56) and Austria (St80). Indoor concentrations in Austria averaged 0.6 pCi/L and ranged from <0.05 to 5.2 pCi/L; those in Sweden generally were in the same range. Some much higher levels in Swedish houses were found associated with low ventilation rates and concrete that contained alum shale (Sw80).

Radon-222 concentrations measured in July-August were only slightly above the March measurements in the CE and NE buildings, but much higher in the ER building. Averages were as follows outside the building and inside on the floor nearest the ground:

Location	March	July-August
outside	0.1 pCi/L	0.2 pCi/L
CE basement	0.6	0.8
NE ground floor	1.2	1.4
ER basement	0.5	3.2

The increases in the CE and NE buildings may be associated with the increased levels in outside air, but the large increase in the ER building must be due to another cause.

Within each building, the highest concentration of Rn-222 was consistently in the story nearest the ground, as summarized in Table 2. Levels in upper floors of the same building were similar. The highest concentrations in buildings are commonly found nearest the ground, particularly in basements (Ge80). This observation gives support to the hypothesis that the main source of RN-222 in building air is the flux from the ground beneath the building. In the NE building, the elevated Rn-222 concentration on the ground floor, which does not have the high-radium concrete blocks used on the upper floors, suggests that these blocks are not the major source of RN-222.

The measurements on 7-28 (see Table 1) show the decrease of Rn-222 levels outside between 7:30 and 11:30 A.M. This is consistent with previous observations of diurnal fluctuations that result in a gradual increase at night to a maximum near sunrise associated with stable atmospheric conditions and possibly inversions during the night, and rapid decrease thereafter (NC75). The changing indoor Rn-222 concentrations at higher levels paralleled those outside. Some of the reduction of Rn-222 concentrations indoors may be due to greater air turnover as more persons used the building in the course of the morning. The measurements on the morning of 7-30, undertaken on an overcast day with relatively stable atmospheric conditions and light rain beginning toward the end of the study period, showed relatively uniform Rn-222 levels outdoors and only a slight decrease in the building.

The highest exhalation rates were found at cracks and seams in the concrete floors of the buildings, and at one location in the ground just outside the ER building, as shown in Table 3. In general, the exhalation rates were several thousand pCi/m^{2}hr from soil and several hundred pCi/m^{2}hr from the concrete blocks. The flux densities at the floors nearest the ground were much less where the floor was continuous than at cracks or seams. The exhalation rates from poured concrete walls in the CE and NE buildings, which have only ordinary Ra-226 contents, were between 10 and 100 pCi/m^{2}hr.

A survey of exhalation rates from soil reported an average value of 1,500 pCi/m^{2}hr (0.75 atoms/cm^{2}sec) and values that ranged approximately three-fold above and below this average in the continental United States (Wi72). Considerable fluctuation can be caused by changes in air pressure and soil moisture content (Co81). The values in Table 3 for the soil near the CE

Table 2

Average Rn-222 Concentrations In Building and Outside Air

Location	Radon-222 concentration, pCi/L				
	Outside	Basement	1st floor	2nd floor	3rd floor
CE building	0.1 (6)*	0.7 (8)	0.4 (2)	0.5 (4)	0.3 (2)
NE building	0.2 (11)	---	1.3 (11)	0.7 (10)	0.8 (4)
ER building	0.2 (5)	2.4 (7)	2.0 (5)	1.8 (4)	---

---

* Number of measurements in parentheses

Table 3

Radon-222 Flux Density, pCi/m^2hr

Date, 1981	CE Building			NE Building			ER Building		
	ground	floor	wall	ground	floor	wall	ground	floor	wall
9-11				3,430(E) 4,360(N)	22,000(C) 48,300(S)	580(W)			
9-15				10,500(N)		270(E)			
9-18							18,300(NE) 5,000(SW)	8,270(S) 6,170(S)	75(W) 220(E)
9-25	2,100(SE) 2,050(SW)	4,630(C) 8,380(C)	10 14						
9-28	2,040(SE) 2,150(SW)			3,820(E) 6,790(N)					
9-29							18,600(NE) 1,840(SW)		
10-16						520(W) 360(E)			15(W) 210(E)
10-23					340 720			430 380 1,900(C)	
11-20		220 680				103 84			

Notes:
1. Letters in parentheses for ground and wall samples show direction relative to building; for floor samples, (C) or (S) indicates measurement over crack or seam, respectively.
2. Samples were collected for 3-day periods, beginning on indicated date.
3. The wall samples labelled (E) and (W) were for high-radium concrete blocks; all others were for poured concrete.

building are near the cited average and were reproducible. Flux densities at some of the other soil locations, however, were relatively high and differed for successive 3-day sampling periods by factors between 2 and 3.

Flux densities between 30 and 650 pCi/m^{2}hr (0.8 to 18 attocurie/cm^{2}sec) had been measured on cellar floors in New Jersey, where the Ra-226 concentration in soil averaged approximately 1 pCi/g (Ge80). The flux densities measured on unbroken surfaces at the ground floors in the NE and ER buildings were toward the upper end of this range. The high values above cracks or seams undoubtedly are higher still relative to the area of the breaks because most of the area of measurement was unbroken floor. As a result, the flux density for the floor as a whole is higher than on unbroken floor, and may reach the exhalation rate from the soil beneath the building (Co81).

Application of the reported ratio of 20 pCi/m^{2}hr (0.005 pCi/m^{2}sec) per Ra-226 concentration of 1 pCi/g in 10-cm-thick concrete (UN77) indicates that most of the exhaled Rn-222 is from beneath the floor. This ratio also gives the order of magnitude of the observed values for the flux density at poured concrete and concrete block walls if their Ra-226 contents are 1.5 and 19 pCi/g, respectively.

The emanation fraction based on the flux density measured for four concrete blocks in the walls averaged 3.0 percent (see Table 4) and ranged from 0.5 to 5.7 percent. The calculation is based on an average Ra-226 daughter concentration of 19 ± 1 pCi/g measured in four blocks that had been removed from walls in these buildings (see Table 5), and the average weight and dimensions determined with these blocks. Note that all of these blocks are hollow. The loss of 3 percent of the Rn-222 by exhalation would increase the Ra-226 concentration to 19.6 pCi/g, consistent with the direct Ra-226 measurement (Table 5) of 20 pCi/g.

For comparison, the emanation fraction for two blocks from the EA building that had been crushed was 3.9 percent, as shown in Table 4. These average values of 3.0 and 3.9 percent are above the range of 0.2 to 1.5 percent observed for 11 samples of Polish concrete that contain slag (Pe80). The emanation fraction measured in 116 samples of ordinary concrete in Hungary, however, ranged from 15 to 60 percent, with a log-mean value of 30 percent (To80). Agreement between the Ra-226 and radium-progeny concentrations in 4 concrete blocks measured here supports a smaller emanation fraction.

The radioactivity levels in surface soil just outside the three buildings, shown in Table 6, were relatively uniform. The overall Ra-226 average of 1.5 pCi/g is above the local average of 1 pCi/g, and the range of 1.0 to 2.6 pCi/g is at the upper end of values between 0.4 and 2.6 pCi/g found in the Atlanta area (Ka79). The measured terrestrial exposure rates listed in Table 6 are consistent with the exposure rates calculated for the radionuclide concentrations. These exposure rates were measured near the soil sampling locations but represent a much larger area, thus providing a measure of confidence that the few samples represent radionuclide levels near the buildings. The terrestrial exposure rate was highest at the location of the highest Rn-222 flux from soil, but the much higher flux density was not

Table 4

Emanation Fraction of Rn-222 from Concrete Blocks

Block	Exhalation rate, pCi/m^2hr	Emanation fraction, %
NE (W)	546 (2)[a]	5.7[b]
NE (E)	316 (2)	3.3
ER (W)	45 (2)	0.47
ER (E)	217 (2)	2.3
EA	---	3.9[c] (3)

---

[a] Number of replicate measurements is indicated in parentheses.

[b] Complete exhalation from one side of a block would be 19 pCi/g x 0.95 g/cm^3 x 7.0 cm x $10^4 cm^2/m^2$ x 0.00755 hr^{-1} = 9,450 pCi/m^2hr.

[c] Amount of Rn-222 in air per weight of block, relative to Ra-226 concentrations of 19 pCi/g.

Table 5

Radionuclide Content of Concrete Blocks

Building	Sampling date	Radioactivity, pCi/g Ra-226	Ra-226 progeny	Ra-228 progeny	K-40
NE	1981	22	19	0.9	9.9
ER	1981	21	18	1.2	9.7
EA	1979	18	17	1.0	7.0
		--	21	1.0	7.7
NE	1979	--	21	1.0	9.4
Average		20	19	1.0	9.1

Note: The last two sets of values are from Ka79, Table 4; block EA was reanalyzed in this study as indicated.

Table 6

Radium-226 Concentrations in Soil Outside Buildings and Confirmatory Exposure Rate Measurements

	CE Building	NE Building	ER Building
Radionuclide concentration, pCi/g			
Ra-226	1.5 ± 0.4*	2.0 ± 0.6	1.4 ± 0.4
Ra-226 progeny	1.0 ± 0.2	1.3 ± 0.3	1.0 ± 0.2
Ra-228 progeny	1.3 ± 0.1	1.4 ± 0.0	1.3 ± 0.3
K-40	12.2 ± 1.3	12.8 ± 0.7	13.0 ± 1.9
Terrestrial exposure rate, μR/hr			
calculated†	7.7 ± 1.2	8.7 ± 0.5	7.9 ± 1.6
measured**	8.4 ± 1.1	8.5 ± 1.2	9.8 ± 1.1

---

* ± values are deviations from mean of duplicate measurements

† Calculated on basis of

$$I = 1.90\ C_{\text{Ra-226 progeny}} + 2.82\ C_{\text{Ra-228 progeny}} + 0.179 C_{\text{K-40}}$$

where C is concentration in pCi/g

** Measured with pressurized ionization chamber; 3.8 μR/hr cosmic-ray exposure rate has been subtracted proportional to the incremental exposure rate above the average. The reported ratio of 2,000 pCi/m^{2}hr (0.5 pCi/m^{2}sec) per soil Ra-226 concentration of 1 pCi/g (UN77) is consistent with observed values except for the highest exhalation rate near the ER building. The Ra-226 level of only 1.8 pCi/g at the surface suggests that the much higher Rn-222 flux density must be attributed to elevated Ra-226 concentrations well below the surface or freer movement of radon through the soil at that location.

Five of the six ventilation measurements obtained on each of the two lower floors in the three buildings yielded rates near 1 per hour, as shown in Figure 1. These rates of exchange of inside with outside air are consistent with the previously reported range in buildings (Ha73). The very high turnover rate for the first floor of the CE building was not used in calculations because the nearby open windows and doors during the ventilation measurement were closed when Rn-222 concentrations had been measured.

The Rn-222 radioactivity level in a structure can be computed from the following balancing of Rn-222 sources from the ground, building materials, and outside air against radioactive decay and ventilation:

$$\frac{d\,C_{Rn}}{dt} = \frac{S_m I_m}{V} + \frac{S_g I_g f_w}{V} - \lambda_{Rn} C_{Rn} - \lambda_a (C_{Rn} - C_o) \qquad (1)$$

if $\frac{d\,C_{Rn}}{dt} = 0$ (equilibrium) and $\lambda_a >> \lambda_{Rn}$ (normal ventilation)

$$C_{Rn} = C_o + \frac{S_m I_m + S_g I_g f_w}{\lambda_a V} \qquad (2)$$

$$I_m = 10^4 C_{Ra} \lambda_{Rn} f_e \rho w \qquad (3)$$

In the above equations, the symbols represent the following:

C_{Rn} : inside Rn-222 concentration, pCi/L

C_o : outside Rn-222 concentration, pCi/L

t : time, hr

S_m : material surface, m^2

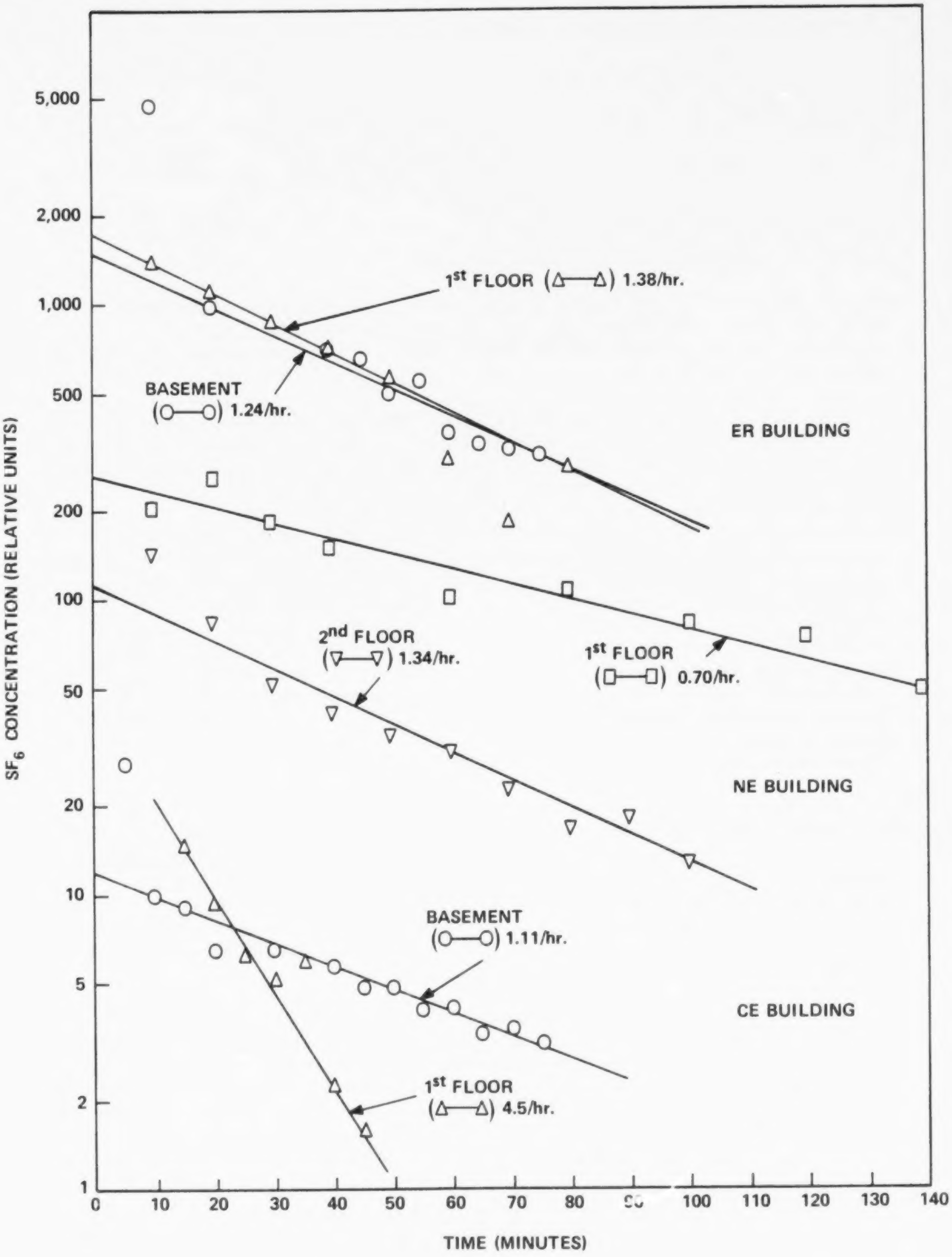
S_g : ground surface, m^2

I_m : inward Rn-222 flux density from material surfaces, pCi m^{-2}hr^{-1}

I_g : inward Rn-222 flux density from ground surface, pCi m^{-2}hr^{-1}

V : building air volume, L

Figure 1



SF$_6$ CONCENTRATION (RELATIVE UNITS)
5,000
2,000
1,000
500
200
100
50
20
10
5
2
1
TIME (MINUTES)
0
10
20
30
40
50
60
70
80
90
100
110
120
130
140
1st FLOOR (△—△) 1.38/hr.
BASEMENT (○—○) 1.24/hr.
ER BUILDING
2nd FLOOR (▽—▽) 1.34/hr.
1st FLOOR (□—□) 0.70/hr.
NE BUILDING
BASEMENT (○—○) 1.11/hr.
CE BUILDING
1st FLOOR (△—△) 4.5/hr.

λ_a : air turnover rate, hr^{-1}

λ_{Rn} : Rn-222 decay rate, 0.00755 hr^{-1}

f_w : fractional Rn-222 diffusion through floor

C_{Ra} : Ra-226 in material, pCi g^{-1}

f_e : exhalation fraction from material

ρ : density of material, g cm^{-3}

w : one-half of wall thickness, cm

In the presence of two types of wall materials, one containing more Ra-226 than the other, the terms $S_m I_m$ can be replaced by $S_m I_m + S_m^* I_m^*$, where the unstarred and starred terms refer to the low-radium and high-radium materials, respectively.

The building dimensions, Rn-222 flux densities and other data in Table 7 were used to compute Rn-222 concentrations in building air. The value of V applies to the entire building. The values of S_m refer to concrete and brick surfaces, including outside walls, inside walls, and floors that are not unusually elevated in Ra-226 concentrations, whereas S_m^* refers to the surfaces of concrete blocks with elevated Ra-226 content. The area of basement walls facing soil was added to the floor area for S_g. The averages of the pertinent measured flux densities in Table 3 were used for I_m^* at the NE and ER buildings and for I_m at all three buildings. The average flux measured in soil near the building was used for I_g in preference to the values measured on the lowest building floor. The differences between measurements on unbroken floor and over cracks or seams were so large that it was assumed that all of the Rn-222 from the ground entered the building through passages through or around the floor (i.e., $f_w = 1.0$). The air turnover rate for a building is the average of the slopes of the pairs of lines drawn through the data in Figure 1.

The Rn-222 concentration in tap water was 1.0 ± 0.6 pCi/L. This value indicates that water use is not a significant source of Rn-222 in building air. At an estimated hourly water use of 1,000 litres, building volumes in excess of 1 x 10^7 liters (see Table 7), and an air turnover rate of approximately 1 per hour, the concentration of Rn-222 in building air from water would be only 1 x 10^{-4} pCi/L. This estimate is consistent with the expectation that water from a surface supply would contribute only minor amounts of Rn-222 to building air (EO79).

The results of these calculations, shown in Table 8, suggest that Rn-222 emanating from the concrete blocks high in Ra-226 has a concentration of 0.1 and 0.2 pCi/L in the NE and ER buildings, respectively. This contribution would approximately double the concentration due to outside air. The concrete materials at the usual lower Ra-226 level in the three buildings contribute an extremely small fraction of the total Rn-222 in building air. The major source of Rn-222 in building air appears to be Ra-226 in soil beneath the

Table 7

Factors for Calculating Rn-222 Concentrations in Building Air

Building dimensions	CE Building	NE Building	ER Building
plan, m^2	40 x 16 + 27 x 4 + 14 x 11 (basement)	69 x 27	65 x 29
height, m	5.5 + 2 x 3.6 + 3.0	3 x 3.0	3.7 + 2 x 3.0
V, m^3	12,000	17,000	18,000
S_m, m^2	16,000	15,000†	11,000
$S_m{}^*$, m^2	-0-	8,500	17,000
S_g, m^2	1,100	1,900	2,100
w_o, g cm^{-2}	16.1 inside 39. outside	7.0	7.0
Radon exhalation and concentration			
I_g, pCi $m^{-2}hr^{-1}$	2,100	5,800	10,900
I_m, pCi $m^{-2}hr^{-1}$	53	53	53
$I_m{}^*$, pCi $m^{-2}hr^{-1}$	-0-	280	280
C_{Ra}, pCi g^{-1}	(1.5)	(1.5)	(1.5)
C^*_{Ra}, pCi g^{-1}	-0-	19	19
Air turnover			
λ_a, hr^{-1}	1.1	1.0	1.2

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* material with elevated Ra-226 content

† does not include outside panel walls

() assumed value

Table 8

Calculated Rn-222 Concentrations in Building Air, pCi/L

Source of Rn-222	CE Building	NE Building	ER Building
Outside air	0.1	0.2	0.2
Ra-226 in ground	0.2	0.6	1.1
Ra-226 in building material - 1.5 pCi/g	0.06	0.05	0.03
- 19 pCi/g	--	0.1	0.2
Total	0.4	1.0	1.5
Measured average	0.6	1.0	2.2

building. The large increase in the Rn-222 concentration in ER building air from March to July may be related to an increased exhalation rate from the ground.

Differences between the measured and computed Rn-222 concentrations shown in Table 8 may be attributable to the following sources of uncertainty: (1) assuming that the Rn-222 flux density at the lowest floor equals that at the ground outside (not considering partial retention of radon by the floor or possibly higher flux rates from soil beneath the building than outside); (2) averaging Rn-222 concentrations for various times and locations; (3) combining concentration and flux density measurements not collected simultaneously or at the same locations; (4) measuring flux density and ventilation rates on only a few occasions; (5) using approximate values for building volumes and surface areas; and (6) obtaining only a few measurements of Ra-226 concentrations and Rn-222 emanation fractions.

SUMMARY AND CONCLUSIONS

In two office/laboratory buildings constructed in part with concrete blocks that contain Ra-226 at the elevated level of 19 pCi/g, average Rn-222 concentrations in air were 1.0 and 2.2 pCi/L, respectively. Radon-222 concentrations averaged 0.6 pCi/L in a control building, and 0.2 pCi/L in outside air. Samples collected simultaneously at comparison locations for 1-min periods showed that Rn-222 concentrations were highest nearest the ground and similar to each other on upper floors.

Radon-222 flux densities averaged 53 pCi/m^{2}hr from poured concrete with normal Ra-226 levels and 280 pCi/m^{2}hr from the concrete blocks with a Ra-226 concentration of 19 pCi/g. The emanation fraction of Rn-222 from the high-radium concrete blocks averaged 3.0 percent on the basis of flux density measurements on site and 3.9 percent for crushed block. The flux density at the floor nearest the ground was several hundred pCi/m^{2}hr over unbroken concrete but one to two orders of magnitude higher over cracks and seams. The flux density over soil near these buildings ranged from 2,000 to 18,000 pCi/m^{2}hr.

According to calculations based on flux density and ventilation rate measurements, the concrete blocks with elevated radium content contributed 0.1 to 0.2 pCi/L to the Rn-222 concentration in building air, equal to the Rn-222 concentration due to outside air during this period. Most of the Rn-222 in air in the two study buildings and the control building is attributed to Rn-222 exhalation from the ground, moving with little or no retention at cracks, seams, and other passages through the concrete. The main differences in building air Rn-222 concentration among the three buildings appear to be due to differences in exhalation rates from the ground.

The calculated Rn-222 concentrations in air in two of the three buildings were one-third below the measured values. Unless a source of Rn-222 has been overlooked, these differences may arise from basing comparisons on averages of relatively few measurements with large differences in flux densities and concentrations. Assuring better comparability would require many more measurements, obtained simultaneously.

Despite the relatively elevated Ra-226 concentrations in the concrete blocks and the extensive use of these blocks in the two buildings, the Rn-222 and inferred WL values in building air were low and the blocks apparently contributed only a small fraction of these. At a typical ratio of 0.004 WL per Rn-222 concentration of 1 pCi/L in building air, the highest average value was 0.009 WL, and the value due to the high-radium concrete blocks was 0.0008 WL. Unless the ventilation rate is drastically reduced and, at the same time, the emanation fraction of the blocks increases greatly, Rn-222 levels from the blocks would not reach limiting levels in the two buildings. The magnitude of the emanation fraction and its dependence on factors such as pressure, temperature, and humidity changes is probably the crucial point in predicting Rn-222 exhalation from building material, and requires considerable additional study.

Acknowledgments: We thank Cornelia Jackson for collecting and measuring the July samples of Rn-222 in air, Jacqualyn Gasper and John Oliver for measuring ventilation rates, and Dr. John Carden for use of the gas chromatograph.

REFERENCES

Bo77 Boothe G. F., 1977, "The Need for Radiation Controls in the Phosphate and Related Industries," Health Phys. 32, 285.

Ca75 Caruthers L. T. and Waltner A. W., 1975, "Need for Standards for Natural Airborne Radioactivity (Radon and Daughters) Concentration in Modern Buildings," Health Phys. 29, 814.

Co81 Collé R., Rubin R. J., Knab L.T., and Hutchinson J.M.R., 1981, Radon Transport Through and Exhalation from Building Materials: A Review and Assessment, NBS Technical Note 1139.

Ea75 Eadie G. G., 1975, "Radioactivity in Construction Materials, A Literature Review and Bibliography," US EPA Rept. ORP/LV-75-1.

Ge80 George A.C. and Breslin A.J., 1980, "Distribution of Ambient Radon and Radon Daughters in New York-New Jersey Residences," The Natural Radiation Environment III, T.F. Gesell and W.M. Lowder, eds., US DOE Rept. CONF-780422, p. 1237.

Gu80 Guimond R.J. and Windham S.T., 1980, "Radiological Evaluation of Structures Constructed on Phosphate-Related Land," The Natural Radiation Environment III, T.F. Gesell and W.M. Lowder, eds., US DOE Rept. CONF-780422, p. 1457.

Ha73 Handley T.H. and Barton C.J., 1973, "Home Ventilation Rates: A Literature Survey," US DOE Rept. ORNL-TM-4318.

Ha78 Harley J.H., 1978, "Radioactivity in Building Materials," Radioactivity in Consumer Products, A.A. Moghissi, et al., eds., Pergamon Press, New York, 336.

Ha78a Harley J. H., 1978, "Radon-222 Measurement," Regional Baseline Station, Chester, NJ, US DOE Rept. EML-347.

He79 Hendricks, D.W., 1979, "Elemental Phosphorus Plants," US EPA Office of Radiation Programs, Las Vegas Facility, Las Vegas, NV, unpublished memorandum.

Hu56 Hultqvist B., 1956, "Studies on Naturally Occurring Ionizing Radiations, with Special Reference to Radiation Doses in Swedish Houses of Various Types," Kungliga Svenska vetenskapsakademiens hadlingar, fjarde serien, Band 6, nr 3, Stockholm.

Ka79 Kahn B., Eicholz G.G., and Clarke F.J., 1979, "Assessment of the Critical Populations at Risk Due to Radiation Exposure in Structures," Report to Office of Radiation Programs, US EPA, NTIS No. PB 81-103764.

Le81 Lepman S.R., Boegel M.L., and Hallowell C.D., 1981, Radon: A Bibliography, USDOE Rept. LBL-12200.

Ll78 Lloyd L., 1978, Montana Dept. of Health and Environmental Sciences, Helena, Montana, personal communication.

Ma78 Maxwell R., Belvin E., and Reed R., 1978, TVA, Muscle Shoals, Alabama, personal communication.

Mo76 Moeller D. W. and Underhill D.W., 1976, "Final Report on Study of the Effects of Building Materials on Population Dose Equivalents," Harvard School of Public Health, Boston, Mass.

NC75 National Council on Radiation Protection and Measurements, 1975, "Natural Background Radiation in the United States," NCRP Rept. No. 45, Washington, D.C.

Pe80 Pensko J. and Stpiczynska Z., 1980, "Emanating Power of Rn-222 Measured in Building Materials," The Natural Radiation Environment III, T.F. Gesell and W.M. Lowder, eds., US DOE Rept. CONF-780422, p. 1407.

Pe78 Peterson B.H., 1978, Idaho Dept. of Health and Welfare, Boise, Idaho, personal communication.

OE79 OECD Nuclear Energy Agency, 1979, "Exposure to Radiation from the Natural Radioactivity in Building Materials," Organization for Economic Cooperation and Development, Paris, France.

Ta80 Task Force on Radon in Structures, 1980, Report of the Task Force on Radon in Structures, U.S. Radiation Policy Council RPC-80-002.

To80 Toth A., Feher I., Lakatos S.N., Koszarus L., and Kezthelyi B., 1980, "Distribution of Natural Radioactive Isotope Concentrations and Exhalation Factors Measured on Concrete and Brick Samples Produced in Hungary," The Natural Radiation Environment III, T.F. Gesell and W.M. Lowder, eds., US DOE Rept. CONF-780422, p. 1396.

Tr79 Travis C.C., Watson A.P., McDowell-Boyer L.M., Cotter S.J., Randolph M.L., and Fields D.E., 1979, "Natural and Technologically Enhanced Sources of Radon-222, Nucl. Safety 20, 722.

UN77 United Nations Scientific Committee on the Effects of Atomic Radiation, 1977, "Sources and Effects of Ionizing Radiation," United Nations, New York.

US80 U.S. Radiation Policy Council, 1980, "Report of the Task Force on Radon in Structures," Rept. RPC-80-002.

Wi72 Wilkening M.H., Clements W.E., and Stanley D., 1972, "Radon-222 Flux Measurements in Widely Separated Regions," The Natural Radiation Environment II, J.A.S. Adams et al., eds., AEC Rept. CONF-720805.

ENVIRONMENTAL RADON INVESTIGATIONS IN SWEDEN

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ABSTRACT

In Sweden, radon daughter concentrations of 200-800 Bq m^{-3} (0.05-0.2 WL) are known to occur in some buildings constructed of areated concrete which have been manufactured from alum shale, a Cambrium, uranium-rich black shale. As a result of these alarmingly high levels, the Swedish government set up, in 1979, a Commission whose task is to initiate research around the problem of radon, and to recommend remedial measures against natural radiation in dwellings.

Based on the recommendations of the Commission, the Government proposed the following measures:

- the introduction of provisional limits for permitted radon daughter concentrations in dwellings. These limits are 400 Bq m^{-3} (0.11 WL) for existing buildings and 70 Bq m^{-3} (0.02 WL) for new development.
- the use of specific building techniques when developing areas with high soil gas radon contents.
- an immediate search for all buildings constructed of alum shale based on aerated concrete.

About 300,000 houses in Sweden are constructed entirely or partially of aerated concrete containing alum shale. During 1980, radon daughter levels have been measured in 20,000 of these houses by Local Health Authorities. The first results show that about 14% of the measured houses have radon daughter levels exceeding 400 Bq m^{-3}. In about 2% of the investigated houses the value exceeded 1,000 Bq m^{-3} (0.27 WL), and maximum values of 4,000 to 9,000 Bq m^{-3} (1.1 -2.4 WL) have been measured in some houses.

Radon daughter levels exceeding 800 Bq m^{-3} are not caused entirely by radon emanating from building materials. A contribution of radon from the ground is necessary. Most of the houses with values exceeding 800 Bq m^{-3} have proved to be sited on ground containing alum shale, uranium-rich granite or uranium-rich pegmatite or on eskers. Many thousands of houses in Sweden are built in such situations.

The measurements so far obtained indicate that the problem of radon penetrating into buildings from the ground is a far greater problem than that of radon emanating from building materials. Geological and geophysical investigations of risk areas are of the utmost importance if all new development is to comply with the provisional limits for radon daughter levels proposed by the Radon Commission.

INTRODUCTION

Approximately 300,000 dwellings in Sweden are constructed, either partially or entirely, of radioactive aerated concrete. This concrete is manufactured from alum shale, a uranium-rich (U 50-350 ppm), Upper Cambrian, black shale which is sometimes sufficiently rich in organic matter that it may be used as fuel. Alum shale occurs as a horizontal layer which varies in thickness from 2 - 75 metres. The distribution of the alum shale occurrences in Sweden is shown in Fig. 1.

The manufacture of alum shale base aerated concrete dates back to 1928. It was known at the time, that the concrete and the alum shale from which it was produced, had high uranium contents, but it was not until during the 1950's that the suitability of alum shale as a building material was questioned (Hultqvist, 1956).

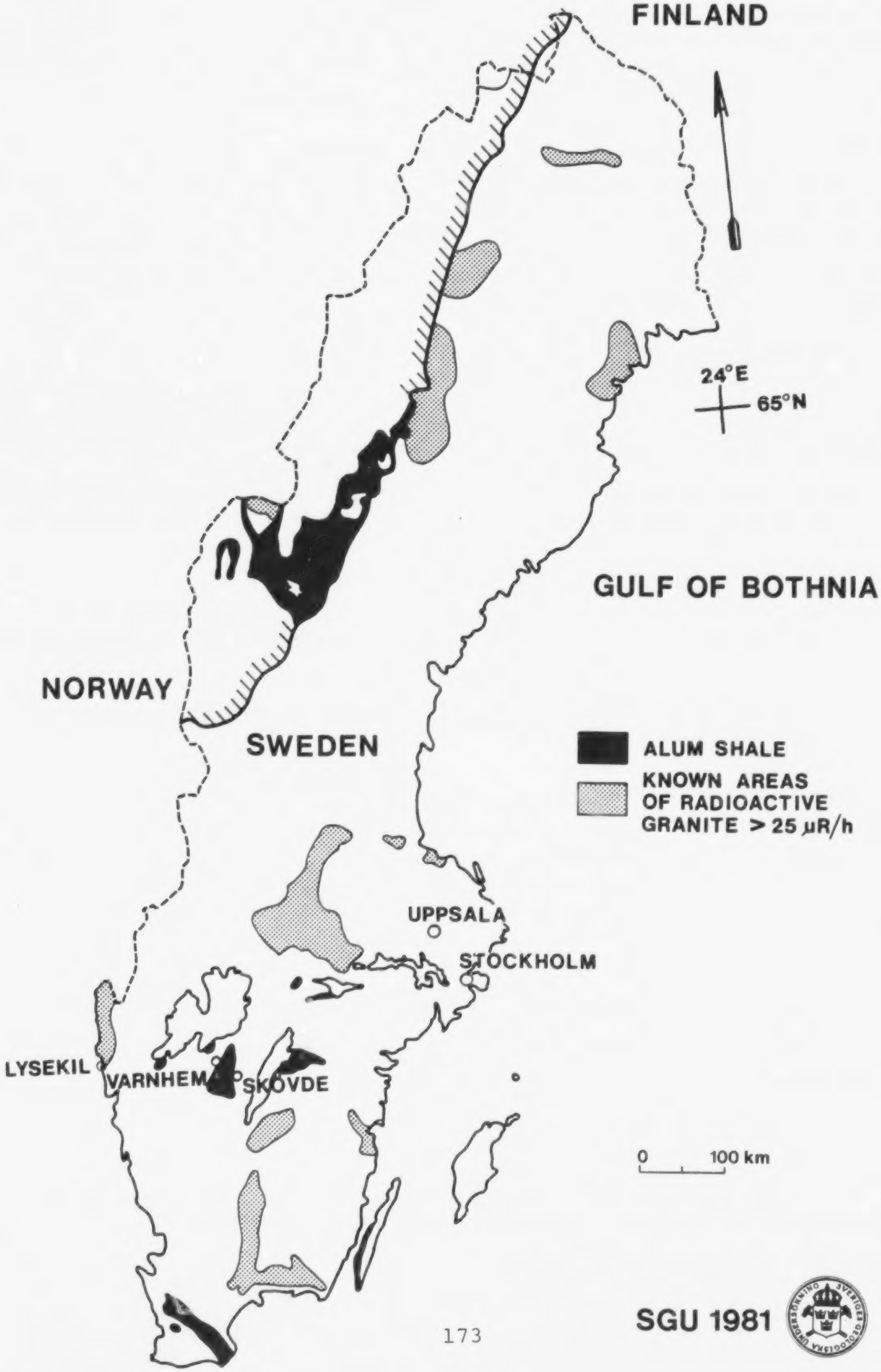
During the 1970's, high indoor radon levels were measured in houses built of alum shale based aerated concrete, but manufacture of this material continued until 1975. Indoor radon measurements made by the National Institute of Radiation Protection during the 1970's revealed that radon levels in houses increased in connection with energy saving measures. Houses were being made more air-tight and rates of ventilation were being lowered. In some of the houses investigated by the Institute, radon levels of 200 - 800 Bq m^{-3} (Becquerel per cubic metre) were encountered (Swedjemark, 1977, 1978 a and b, 1980).

A new dimension to the radon problem became apparent in 1978 when the National Institute of Radiation Protection measured radon levels in houses built of normal materials on a foundation of alum shale tailings. The measured levels were found to be between 400 and 1,600 Bq m^{-3} (Swedjemark, et al 1979).

RADON COMMISSION

In February 1979, the Swedish Government appointed a Commission to investigate measures against radiation hazards in buildings. The Commission was also instructed to initiate a search for all such buildings, and to promote research around the radon problem. The Commission is assisted by various experts, for example, from the National Institute of Radiation Protection, the National Board of Health and Welfare, the National Board of Urban Planning, the Geological Survey of Sweden, the building industry and the Swedish Association of Local Authorities. A preliminary report from the Commission was presented in May 1979 (Radonutredningen 1979). The report contains proposals on: provisional limits for the maximum radon daughter levels in dwellings; maximum permitted radium content in building materials and gamma radiation from these materials; searches for houses with high radon daughter levels; and research programs.

Figure 1


FINLAND
24°E
65°N
GULF OF BOTHNIA
NORWAY
SWEDEN
■ ALUM SHALE
▨ KNOWN AREAS OF RADIOACTIVE GRANITE > 25 µR/h
UPPSALA
STOCKHOLM
LYSEKIL
VARNHEM
SKÖVDE
0 100 km
SGU 1981

A further proposal made by the Commission in its report was that maps, termed GEO-radiation maps, should be prepared to show the distribution in Sweden of all areas with particularly radioactive rock and soils (alum shale and uranium-rich granites and pegmatites). A full description of these maps is given in Wilson (1981).

LIMITS

Acting on instructions from the Government, the National Board of Health and Welfare and the National Board of Urban Planning in consultation with the National Institute of Radiation Protection have issued provisional regulations based on the proposals presented by the Radon Commission (Socialstyrelsen 1980, Statens planverk 1980). In short, the regulations are as follows (complete text is given in Appendix 1.):

Existing houses

400 Bq m^{-3} (0.11 WL) is a provisional action limit for the annual average of the equilibrium concentration of radon. Houses with levels exceeding 400 Bq m^{-3} are declared to be insanitary.

After remedial action or building alterations, the annual average of the equilibrium equivalent concentration of radon shall not exceed 200 Bq m^{-3}.

New development

70 Bq m^{-3} (0.02 WL) is the maximum permitted limit for the annual average of the equilibrium equivalent concentration of radon in rooms which are in continual use.

50 μR/h (micro Roentgen per hour) is the maximum permitted gamma radiation for rooms in continual use.

Building materials

Building materials for use in continuously inhabited constructions shall not have a gamma index or radium index exceeding 1.0. It is also recommended that gamma radiation shall not exceed 100 μR/h for outdoor areas which are in regular use, for example, playgrounds.

The gamma and radium indexes are defined as follows:

$$\text{Gamma index} = \frac{C_K}{10{,}000} + \frac{C_{Ra}}{1{,}000} + \frac{C_{Th}}{700}$$

$$\text{Radium index} = \frac{C_{Ra}}{200}$$

where C_K, C_{Ra} and C_{Th} are the concentrations of potassium-40, radium-226 and thorium-232 expressed in terms of Bq m^{-3} of building material.

70 Bq m^{-3} (0.02 WL), the maximum permitted level for new development, is a functional requirement implying that technical measures may be necessary against all sources of radon gas, for example, radon in the ground, in building materials and in household water supplies. This regulation supercedes the proposal from the Radon Commission that, in areas where gamma radiation in the ground at foundation level exceeds 30 μR/h, houses shall either be constructed "radon safe" or the site investigated to determine whether a "radon safe" construction is necessary.

SEARCHES FOR "RADON HOUSES"

Searches for houses with high radon daughter levels have been in progress since the spring of 1979. Initially the search was aimed at finding houses constructed with alum shale based aerated concrete, but more recently attention has been focused on the search for houses where high indoor radon levels are related to high soil radon concentrations.

The search for houses constructed with radioactive concrete faced certain problems. As house deeds generally contain no information as to the type of concrete used, most houses in the country need to be visited in order to determine whether or not they are constructed of radioactive concrete.

The searches have been the responsibility of the Local Health Authorities. These authorities have relied upon information from householders and measurements of gamma radiation using handborne scintillometers or carborne scintillometer measurements (Wahren et al 1979). The latter method has proved to be very effective permitting control of radiation from the outer walls of 600 to 1,000 houses per day.

The carborne measurements carried out by the Geological Survey of Sweden use a scintillometer with a sodium iodine (NaI) crystal which has a volume of 6,130 cubic centimetres. The instrument is mounted in a car with a shield against radiation from the ground in order to reduce the background radiation contribution (Fig. 2). A follow-up program of measurements of indoor radon daughter levels has commenced for those houses which are judged to have a problem.

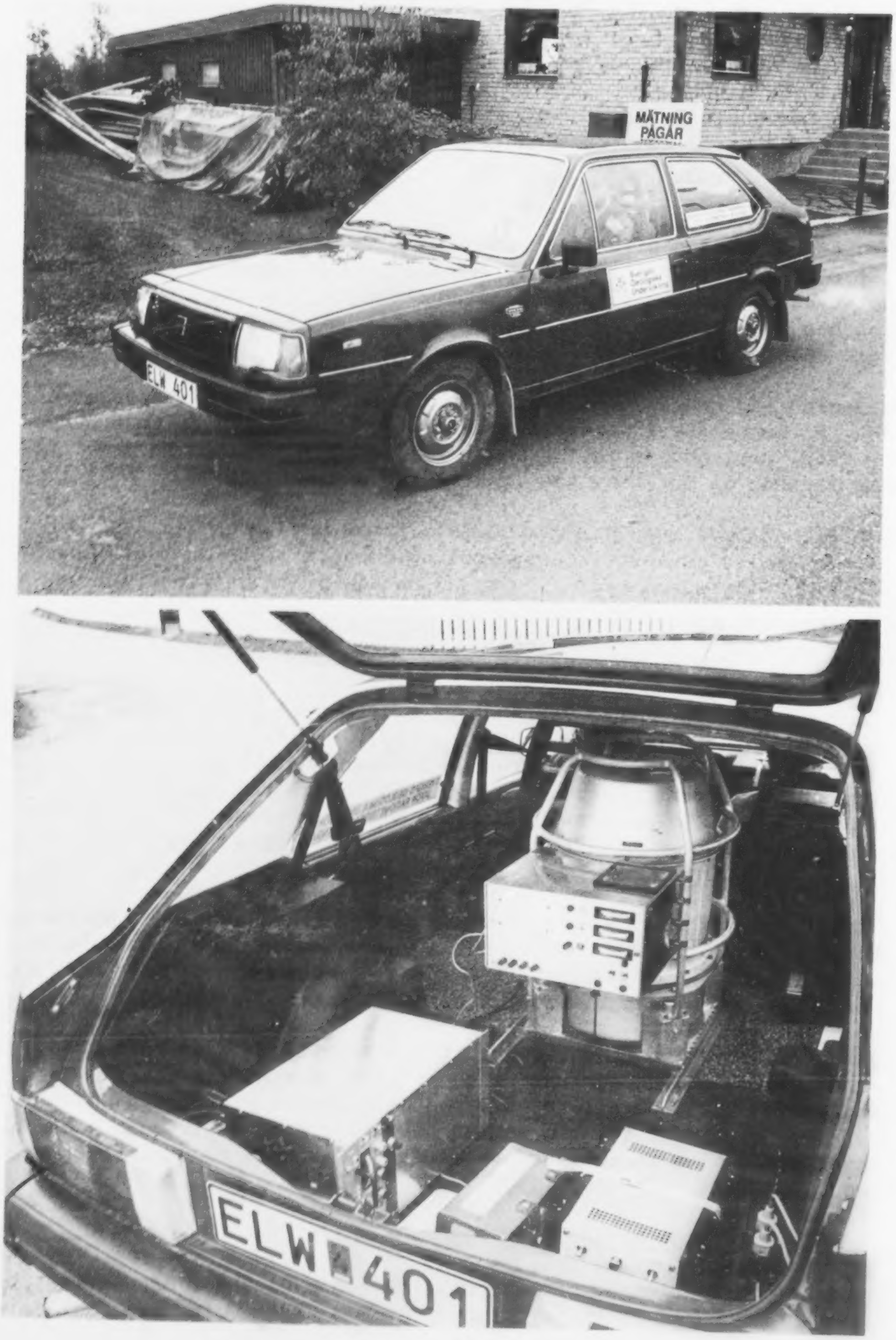
To date (Spring 1981), radon daughter measurements have been performed in approximately 20,000 houses. Most of the measurements have been made using alpha-sensitive film. On a more limited scale, measurements have been made using the filter method or thermo-luminiscence detectors (TLD).

For most of the film measurements, Track Etch films have been used. The films (2 per house) have been used without radon daughter filters. The measuring period has generally been 3 months, summers preferably being avoided. The results of the two measurements for each individual house have been converted to give an average annual radon daughter concentration (Samuelson, 1980).

RESULTS OF INDOOR RADON MEASUREMENTS

The first compilation of measurements of radon daughter concentrations from 5,600 houses built of radioactive aerated concrete gives the following results: 46% of the houses have levels exceeding 200 Bq m^{-3} (0.05 WL), 14%

Figure 2



levels exceeding 400 Bq m^{-3} (0.11 WL) and 2% levels exceeding 1,000 Bq m^{-3} (0.27 WL) (Hildingsson, 1981). A more recent compilation for 12,000 houses gives very similar results: 48% with levels exceeding 200 Bq m^{-3}, 13% levels exceeding 400 Bq m^{-3} and 1% levels exceeding 1,000 Bq m^{-3} (Hildingsson, pers. comm.).

Clear geographical variations in the radon daughter levels are apparent from the results so far obtained. These variations are caused, either by variations in the radium content of the aerated concrete used, or by variations in the soil gas radon concentrations, the latter being closely related to the geology. Variations in the radium content of the aerated concrete are due to the fact that this type of concrete was manufactured in different parts of Sweden using alum shale with widely varying uranium contents.

There is a marked increase in the number of houses with high indoor radon levels in areas where the bedrock consists of alum shale or uranium-rich granites and pegmatites (Table 1). It has been found, however, that sporadic occurrences of high indoor radon levels also occur in areas with normal uranium contents in the bedrock or soils. Enchanced soil gas radon levels have been noted in glacial eskers with normal uranium contents of 3-8 ppm U in the soil and in areas with scattered occurrences in uranium-rich pegmatites.

The initial results of the program of indoor measurements show clearly that the problem of radon emanation from the ground is greater than that from building materials. At a rough estimate, 3,000 - 15,000 houses in Sweden are in need of remedial measures against radon penetration from the ground. Tracing these houses will be difficult. One way would be to carry out indoor measurements in all houses built in areas known to have high uranium contents in the bedrock or soil cover. Tracing of the sporadic ocurrences is however a problem. It should also be borne in mind that exceptionally high soil gas levels exist in houses which are ventilated in such a way that large quantities of air are drawn into the building through cracks, etc. in the foundation or cellar walls.

RESEARCH

A research program, initiated by the Radon Commission, is underway to investigate the relationship between radon concentrations and uranium content in the ground and indoor radon and gamma ray levels. 100 houses with high indoor radon daughter levels from areas of alum shale or uranium-rich granite have been selected for detailed studies of the bedrock, soils, groundwater and building techniques. These houses are not constructed from radioactive aerated concrete. The investigations will include in situ determinations of the U-, Th- and K-content of the rocks and soils using gamma spectrometers, gamma logging, measurements of the soil gas radon content and/or radon emanation from the surface, measurement of radon content in the groundwater and in the drainage layer under the houses, and measurements of radon daughter levels in the houses. The results of the investigations should be available in 1982.

Other research programs are also in progress where various types of remedial measures are being studied, including means of ventilation and the laying of aluminum foil on the floor to prevent radon penetration from the ground.

Locality	Building material	Radon daughter levels, Bq m^{-3}, in the investigated houses and apartments				Geology
		% > 200	% > 400	% > 100	no. houses investigated	
Uppsala (houses)	aerated concrete	3.5	1.0		516	Till, sand, gravel and clay overlying granodiorite and acid to basic volcanics (U in bedrock 1-8 ppm)
Skovde (houses and apartments)	"-	79	42	9[1]	380	Sand, clay and alum shale overlying sandstone, alum shale and limestone (U in alum shale 50-300 ppm)
Stockholm suburb (houses)	"-	65	42	6[2]	278	Till, sand and clay overlying gneisses younger U-rich granites and pegmatites (U in gneiss 2-6 ppm, in granites 10-20 ppm). Well exposed.
Varnhem (houses)	other type of b. material	90	48	22	31	Alum shale rich till, sand and gravel overlying sandstone and alum shale (U in alum shale 50-300 ppm)
Lysekil (houses)	"-	20	10	2	63	Uranium-rich granite, exposed (U content 12-37 ppm)

[1] 6 houses 4,000 Bq m^{-3}
1 house 8,000 Bq m^{-3}

[2] 1 house 6,000 Bq m^{-3}

Table 1. Variations in indoor radon daughter levels reflecting the geology.

SITE INVESTIGATIONS

In order to comply with the regulations concerning radon daughter levels in the new buildings (70 Bq m^{-3}), one needs either to build houses "radon safe", or to know that the soil gas radon content is so low that no indoor problem will arise. In the case of building on ground with enhanced radon concentrations, the type of "radon safe" construction used may be varied according to the actual soil gas radon levels.

The purpose of the site investigations is to determine whether or not the soil gas radon concentration within a planned area will give rise to high indoor levels once the houses are built.

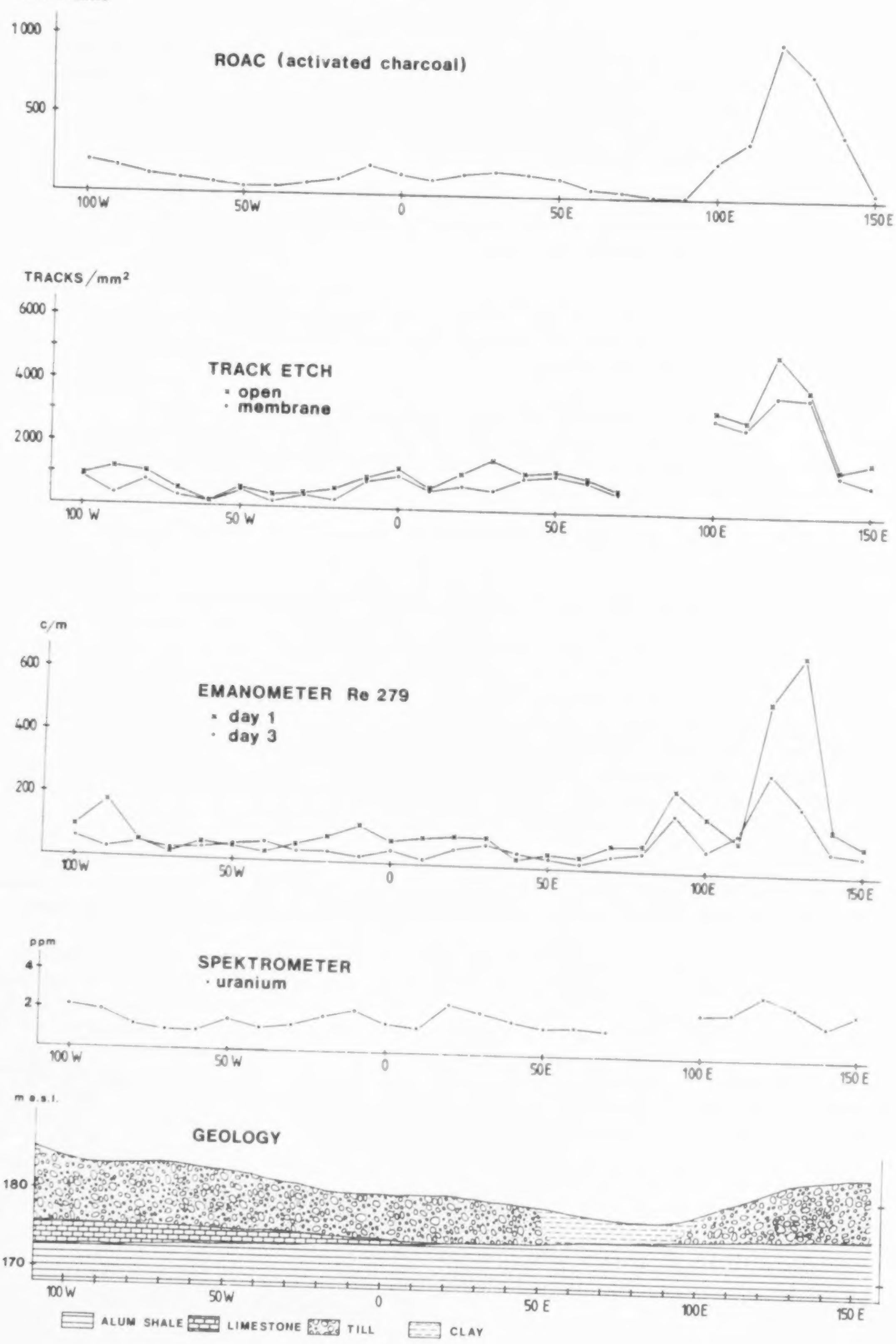
Account must be taken of the changes in the soil gas radon concentrations which will be caused by the actual process of building, for example, lowering of the water table or removal of some of the soil cover so that the foundations lie at a deeper level.

For the site investigations carried out by SGU, soil gas radon measurements are made at a depth of one metre in order to minimize temporary fluctuations caused by variations in wind, temperature and precipitation without increasing the measuring costs to unacceptable levels. In conjunction with the radon measurements, observations are made on soil types, bedrock geology, ground water and gamma radiation both above the ground and in the soil.

The methods used by SGU are ROAC-cups which are filled with activated charcoal (Hambleton-Jones and Smit, 1980). Track Etch alpha sensitive film (Gingrich and Fisher, 1976) and emanometers. The measuring time for ROAC-cups is 5-7 days, and for Track Etch film 3 weeks. These methods were tested during 1980 in a research project initiated by the Radon Commission (Hesselbom et al, 1981). Measurements were performed in both granite and alum shale environments. The ROAC and Track Etch cups gave comparable results, but the emanometer readings varied, compared with ROAC and Track Etch, from area to area probably because varying amounts of atmospheric air were pumped into the instrument. The amount of interference from atmospheric air will depend on the porosity and permeability of the soil in which the measurements are made. The advantage of the emanometer method is that it is quick, and it gives an idea as to whether radon levels in the soil are enhanced or not. Fig. 3 shows the results of test measurements carried out by SGU in an area of alum shale.

The bedrock along the profile consists of alum shale (U 150 - 300 ppm) overlain to the west by limestone (U less than 2 ppm). The soil cover is 5-10 metres thick and consists of clay to sandy till, and in the central part of the profile of clay. The water table lies 2-3 metres above the bedrock surface. In the western part of the profile the till is comprised of limestone fragments, alum shale fragments are absent. This also applies to the upper 4 metres of the till in the eastern part of the profile. Here the content of alum shale fragments increases successively with depth.

Figure 3


ROAC units
1000
500
ROAC (activated charcoal)
100 W
50 W
0
50 E
100 E
150 E
TRACKS/mm²
6000
4000
2000
TRACK ETCH
▪ open
· membrane
100 W
50 W
0
50 E
100 E
150 E
c/m
600
400
200
EMANOMETER Re 279
▪ day 1
· day 3
100 W
50 W
0
50 E
100 E
150 E
ppm
4
2
SPEKTROMETER
· uranium
100 W
50 W
0
50 E
100 E
150 E
m a.s.l.
GEOLOGY
180
170
100 W
50 W
0
50 E
100 E
150 E
ALUM SHALE
LIMESTONE
TILL
CLAY

The radon measurements show that the radon concentration in the soil for the western part of the profile is low, approximately 3,000 to 30,000 Bq m^{-3}. This can be related to the low uranium content of the till combined with the shielding effect of the limestone horizon, the till cover and the high water table all of which prevent radon from the alum shale horizon from reaching the surface. In the eastern part of the profile the shielding effect is no longer present and the soil gas radon content is approximately 120,000 - 160,000 Bq m^{-3}.

The results show a good correlation between the measurements performed with ROAC and Track Etch cups and the emanometer.

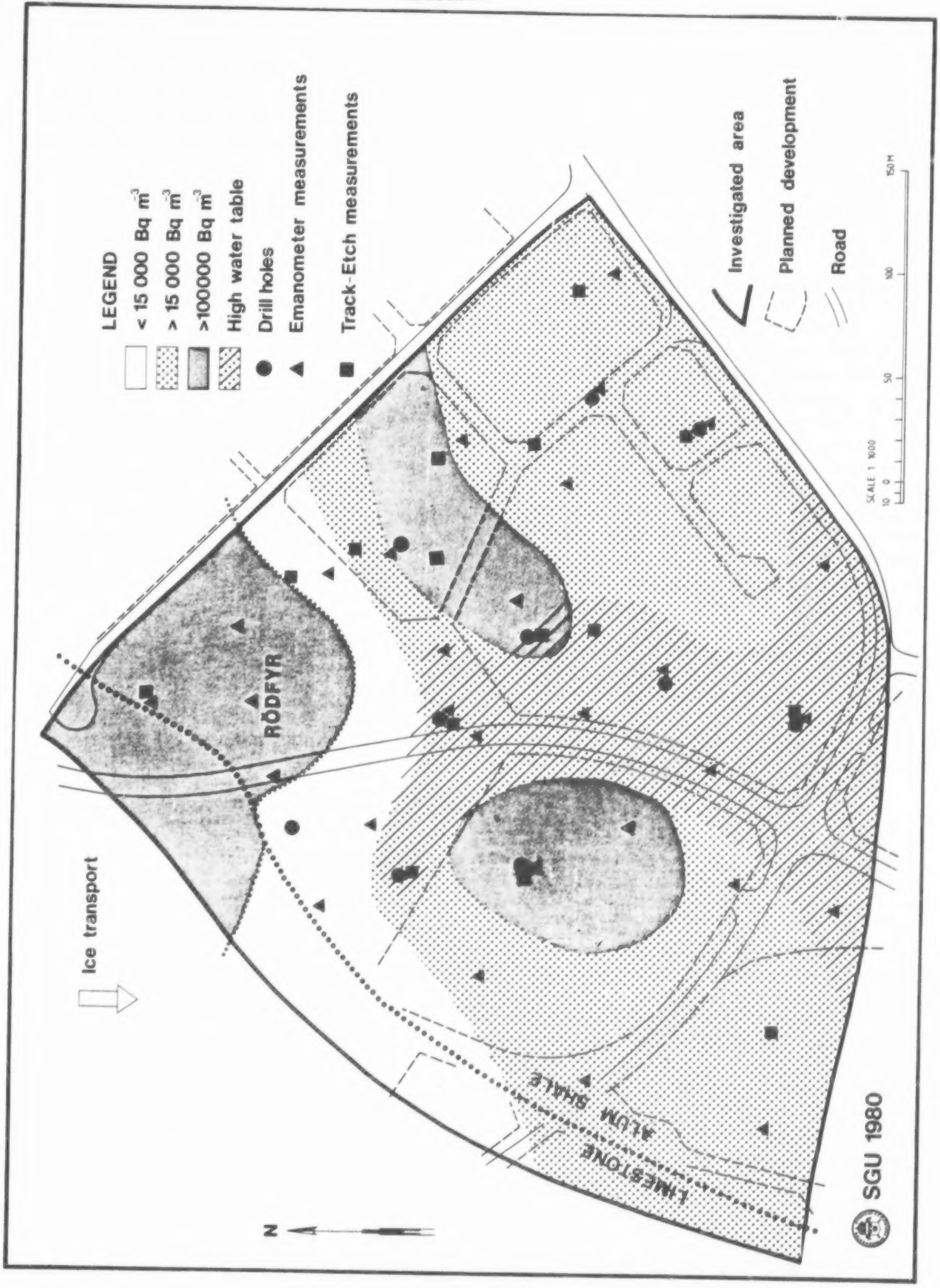
None of the methods used by SGU are calibrated to give absolute concentrations of radon in the soil in Becquerels per cubic metre. Track Etch, however, quote an average value of approximately 6,000 Bq m^{-3} for all soil gas radon measurements performed by their method. Of the measurements carried out by SGU at a depth of one metre in till or gravel containing alum shale, values some 30 to 300 times larger than the average value quoted by Track Etch have been obtained. For till or gravel containing fragments of uranium-rich granite, values 25 to 75 times the average Track Etch value have been measured. In other words, soil gas radon levels in areas of alum shale can be as high as approximately 500,000 - 1,800,000 Bq m^{-3}, and in areas of uranium-rich granites 200,000 - 500,000 Bq m^{-3}. With such high soil gas radon levels, intake of more than 70 litres per hour of soil air (radon content 500,000 Bq m^{-3}) into a house with a ventilation rate of 0.5 air exchanges per hour will be sufficient to produce indoor radon daughter levels exceeding 70 Bq m^{-3}.

Fig. 4 illustrates an area investigated by SGU in the county of Narke, central Sweden. The purpose of the investigation was to determine the radon content in the soil air in an area planned for development. The investigation included radon measurements using both an emanometer and Track-Etch film, gamma-radiation measurements with a handborne scintillometer, and gamma-logging of drill holes.

The bedrock of the area consists of alum shale which is overlain to the west and north by limestone. The bedrock is covered by till and sand. In the eastern part of the area, layers of clay occur interbedded in the sand. The surface of the till lies at 1-3.4 metres depth. In the northern and western parts of the investigated area, the till is dominated by limestone fragments, whereas in the southern and eastern part alum shale fragments dominate. The water table lies at a depth of 0.4-1.1 metres. Tailings of burnt alum shale (rödfyr) have been tipped in the northern part of the area.

The gamma radiation measurements made at ground level gave low readings of 6-11 μR/H, except for the area with rödfyr where the radioactivity is 90-115 μR/h. The gamma-log results also gave low readings of 3-10 μR/h with a slight increase to 14-19 μR/h for soils containing a few fragments of alum shale. It should be noted, however, that the drill holes only penetrated the upper soil layers down to the upper surface of the till. The readings therefore do not apply to the radiation levels of the till.

Figure 4


LEGEND
< 15 000 Bq m^{-3}
> 15 000 Bq m^{-3}
>100000 Bq m^{-3}
High water table
Drill holes
Emanometer measurements
Track-Etch measurements
Investigated area
Planned development
Road
SCALE 1:1000
10 0 50 100 150 M
Ice transport
RÖDFYR
LIMESTONE
ALUM SHALE
N
SGU 1980

The radon measurements were made at the greatest possible depth (0.9 metres for Track Etch and 0.7 metres for the emanometer) but the high level of the water table often prohibited this.

On the basis of the results of the radon measurements, the investigated area can be subdivided into three (Fig. 4). In the north and northwest where the till is dominated by limestone fragments, the radon concentration in the soil gas is normal to slightly raised.

In the southern and eastern parts of the area, where the till is dominated by alum shale fragments, the soil gas radon level is higher than normal, 30,000 - 100,000 Bq m^{-3}. Within the latter area however, lower values were obtained where the water table was very near the surface. The high ground water level hinders the radon gas from the alum shale till from reaching ground level. If, however, the water table is lowered, either naturally or as a result of construction work, the content of radon in the soil gas will increase in these areas. Soil gas radon levels in the areas of tipped rödfyr exceed 100,000 Bq m^{-3}.

In the areas with high levels of soil gas radon, the planned dwelling should be so constructed as to prevent infiltration of radon from the ground into the dwellings.

References

Gingrich, J.E. and Risher, J.C., 1976. Uranium exploration using the Track-Etch method. IAEA-SM-208/19, 213-225.

Hambleton-Jones, B.B. and Smit, M.C.B., 1980. ROAC - a new dimension in radon prospecting. Atomic Energy Board, Pretoria, South Africa. ISBN 0 86960 7081, 24 pp.

Hesselbom, A., Israelsson, S. and Tovedal, H., 1981. Radon in the ground. A study of methods and instruments for determining radon concentrations in the ground. Byggforskningsradet, Report R47:1981 (in Swedish), 75 pp.

Hildingsson, O., 1971. Radon daughter levels in 5,600 houses. Measurements made using alpha sensitive film and filter method. Technical Report, Statens Provningsanstalt, SP Rapp. 1981:27, (in Swedish).

Hultqvist, B. 1956. Studies on naturally occurring ionizing radiations with special reference to radiation doses in Swedish houses of various types. Svenska vetenskapsakademins handlingar, fjarde serien, Band 6, nr 3, Stockholm, Sweden.

Radonutredningen, 1979. Preliminary proposal for measures against radiation hazards in buildings. Report from the Radon Commission, Jordbruksdepartementet, Ds Jo 1979:9 (in Swedish), 114 pp.

Samuelson, I. 1980. Determination of the annual average radon daughter concentration in a house measured with alpha sensitive films. Statens Provningsanstalt, SP A2 601, 8 pp.

Socialstyrelsen, 1980. Statement from the National Board of Health and Welfare with advise and regulations concerning remedial measures against radon in dwellings, September 1, 1980. Socalstyrelsens forfattningssamling, SOSFS (M) 1980:71, 4 pp, (in Swedish).

Statens Planverk, 1980. Svensk Byggnorm 1980 (Swedish Building Code 1980). Statens planverks forfattningssamling 1980:1 (in Swedish).

Swedjemark, G.A., 1977. The ionizing radiation in dwellings related to the building materials. Statens stralskyddsinstitut, Report SSI:1977-004, 1-9.

- 1978a. Radon in dwellings in Sweden. In Proceedings from the Symposium on Natural Radiation Environment III, Houston, Texas, April 23-28, 1978.

- 1978b. The effects of energy saving reduced ventilation on radiation dose in dwellings. Statens stralskyddsinstitut, Report SSI:1978-014, 1-13 (in Swedish).

- 1980. Radioactivity in houses built of aerated concrete based on alum shale. Statens stralskyddsinstitut, Report SSI:1980-14, 1-6.

Swedjemark, G.A., Hakansson, B. and Hagberg, N., 1979. Radiation levels in houses built on wastes from processing of alum shale. Statens stralskyddsinstitut, Report SSI:1979-006, 1-43 (in Swedish).

Wahren, H., Makitalo, A., Persson. T. and Svensson, C.-E, 1979. An attempt to trace 'radon houses' in the municipality of Uppsala. Delrapport 1979, Lanslakarorganisationen i Uppsala lan, 62 p, (in Swedish).

Wilson, C., in press. Regional environmental documentation of natural radiation in Sweden.

Appendix: Summary of provisional regulations and recommendations concerning radiation hazards in buildings in Sweden

1. Directions issued by the National Board of Health and Welfare, in force from September, 1980.

   A building is regarded as insanitary when the radon daughter concentration in rooms in continual use exceeds an annual average of 400 Bq m^{-3}. SOSFS(M) 1980:71 (Socialstyrelsen 1980).

2. Mandatory regulations issued by the National Board of Urban Planning, SWEDISH BUILDING CODE (SBN) 1980, in force from January 1, 1980. National Board of Urban Planning Statute Books 1980:1 (Statens planverk 1980).

New buildings

Buildings shall be so constructed that the annual average concentration of radon daughters in rooms in continual use shall not exceed 70 Bq m^{-3}.

The annual average concentration of radon daughters is determined according to methods laid down by the National Board of Urban Planning in consultation with the National Institute of Radiation Protection.

SBN 36:41

The regulation relating to a maximum permitted indoor radon daughter concentration of 70 Bq m^{-3}, according to 36:41, can require the application of remedial measures against sources of radon in the ground, in household water supplies and in building material.

SBN 31:142

A building shall be so constructed that gamma radiation in rooms in continual use shall not exceed 50 μR/h.

SBN 31:141

A satisfactory level for gamma radiation is achieved if the regulation relating to building materials, 31:143, is applied, and if gamma radiation from the ground is screened by a concrete structure or filling of low radioactive material.

SBN 31:1411

Rebuilding

Buildings shall be so constructed that the annual average concentration of radon daughters in rooms in continual use shall not exceed 200 Bq m^{-3}. Exception is made where the radon daughter concentration cannot be brought below this level by means of increased ventilation and remedial measures according to 31 rebuilding: 14.

SBN 36: Rebuilding: 41

Exception to the regulation quoted in SBN 31:141 may be granted. (That is to say, there are no regulations which apply to lowering of gamma radiation levels in cases of rebuilding.)

SBN 31: Rebuilding: 141

Exception to the regulation quoted in SBN 31:143 regarding existing building materials, may be granted. However, an accessible, strongly radon emanating filling material shall be removed where required in order to conform with the regulation concerning radon daughter concentrations quoted in SBN 36 Rebuilding: 41.

Easily accessible filling may be found against cellar walls, and in the framing of joists and the crawl space.

SBN 31: Rebuilding: 143

Building materials

Building materials used in buildings in continual use shall not have gamma or radium indexes exceeding 1.0.

SBN 31: 143

Filling materials and material used in the drainage layer under the foundations also come under the heading building materials.

SBN 31: 1431

Gamma- and radium indexes are defined as follows:

$$\text{Gamma index} = \frac{C_K}{10,000} + \frac{C_{Ra}}{1,000} + \frac{C_{Th}}{700}$$

$$\text{Radium index} = \frac{C_{Ra}}{200}$$

where C_K, C_{Ra} and C_{Th} are the concentrations of potassium-40, radium-226 and thorium-232 expressed as Bq kg^{-1} of bulding material.

Out of doors

There are no regulations which apply to gamma radiation levels out of doors. However, it is recommended that in areas which are in regular use, for example playgrounds, gamma radiation should not exceed 100 μR/h.

Figure Captions

1. Distribution of the alum shale formation and of areas known to contain radioactive granites.

2. Car and equipment used in the search for houses with aerated concrete.

3. Results of test measurements made in an area of alum shale (after Hesselbom et al 1981).

4. Map showing the results of radon measurements in an area planned for development.

EFFECT OF AIR CIRCULATION
AND DUST REMOVAL ON INDOOR
RADON DECAY PRODUCT CONCENTRATION

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ABSTRACT

The effectiveness of increased room air motion and dust removal in reducing working levels in residences subject to radon intrusion was evaluated in a 90-m^3 chamber under steady-state conditions for air infiltration rates between 0.2 and 0.8/hr. Room-sized electrostatic precipitators and high-efficiency fibrous filters were tested as typical residential air cleaning devices; a portable box fan and a ceiling fan were employed as typical residential air movers. Reductions in working levels from 40 to 90% were measured. The fate of radon decay products was determined by direct measurement. When mixing fans were used, most of the potential alpha energy deposited was plated out on the room surfaces; less than 5% was deposited on the fan blades. Results were compared to a mathematical model based on well-mixed room and good agreement was obtained.

INTRODUCTION

Strategies to control radon decay products in residences can be divided into three types: (1) prevention of radon entry into homes (e.g., caulking, sealing, barrier paints, and judicious selection of building materials); (2) dilution of outside air directly or through heat exchangers; (3) removal of radon or radon decay products from indoor atmosphere by various air treatments. Historically, strategy (2) has been most widely used, perhaps inadvertently. In recent years, it has become less acceptable because of the high cost of energy for space heating and cooling.

The present study was directed to strategy (3), although no attempt was made to cleanse the air of radon gas. The treatment methods evaluated included the use of room-sized electrostatic precipitators, high-efficiency air filters, and air mixing by fans. Because much of the data collected has not been analyzed, this paper should be considered a first reporting of on-going research. More complete results will be available in three months.(1)

Very little has been published on the use of methods to remove airborne radon decay products. The experimental work that has been reported, concerned primarily with the reduction in radon decay product concentrations in mines,(2-8) has only limited application to control in residences because of the higher dust concentration, higher air infiltration rates, and higher humidity found in mines.

EXPERIMENTAL PROCEDURES

Experimental studies were conducted in a 90-m^3 chamber, about the size of a large room in a house. Special attention was given to making this chamber as much like a residence as possible, although certain differences are noteworthy. The chamber's 4-m high ceiling is not characteristic of houses, nor are its painted metal walls. We do not believe, though, that these differences are important. Transient effects in houses, such as changes in air infiltration or radon intrusion rates with time, however, are undoubtedly important, but we made no attempt to simulate this behavior. In fact, to facilitate interpretation of our experimental results, we held all parameters within our control constant, and no data were retained for analysis until steady-state conditions had been reached.

Air Infiltration

The natural air infiltration rate into our experimental chamber was very low; from the decay rate of a CO tracer gas,(1) we measured about 0.015/hr. Because we wanted to hold the air infiltration rate constant at various settings and still simulate air infiltration into a room of a house, a blower was installed outside the chamber to exhaust air from it. (See Figure 1.) No inlet air port was provided and, thus, incoming air was forced to infiltrate through the cracks and pores in the walls or the seals around the door and windows. Pressure in the chamber was never more than 3-mm Hg below barometric pressure and was usually less. The exhaust airflow rate was measured with a calibrated venturi flowmeter, and the rate controlled with a butterfly damper. The air infiltration rate was equal to the measured airflow rate divided by the chamber volume. The particulate level and size distribution of the infiltrating air was about the same as laboratory air, although somewhat variable, and is reported elsewhere.(1)

Radon Generation

Radon-222 gas was generated by de-emanation of a 100-µCi solution of Ra-226. About 0.2 L/min of prehumidified air flowed continuously through a bubbler containing the radium solution, through a liquid carryover trap and membrane filter, and into a distribution manifold placed on the floor of the chamber to simulate radon emanating from the subsoil and entering through the floor. This is shown schematically in Figure 1. The manifold was constructed from 3.5-cm I.D. plastic pipe in which 0.6-mm holes were drilled. Inasmuch as the holes are much smaller than the pipe diameter, gas is evenly discharged from all holes. These holes face downward and are located on 60-cm centers throughout the chamber. About 5 L/min of dilution air was also introduced into the distribution manifold.

Sampling and Measurement of Radon Decay Products

As shown in Figure 1, sampling ports were provided in the exhaust duct and on one wall of the chamber. Both ports accept 50-mm diameter open-faced filter holders, which minimize aerosol sampling losses. To determine whether these samples would be representative of the contents of the chamber, we made tests in which a tracer gas (CO) was introduced continuously through the distribution manifold. After steady state had been reached, samples were taken from the two sampling ports as well as from eight other locations at various heights in the chamber. All samples were in reasonably good agreement over the range of air infiltration rates we studied.(1) Further verification that the chamber was well mixed can be made from a comparison of samples taken from the exhaust duct and wall port. For 25 pairs of samples, the exhaust duct gave an average of 0.0907 working level (WL) compared to 0.0890 WL from the wall port, about a 2% difference.

Filter samples were counted by alpha scintillating using disposable Ag-activated ZnS phosphors coated on one side of a Mylar film (William B. Johnson & Associates, Inc., Research Park, Montville, New Jersey 07045), which were placed in direct contact with a bare photomultiplier tube and enclosed in a light-tight box. Working level was determined by the modified Tsivoglou method.(9)

MODELING

Because samples taken from various locations in the experimental chamber showed good agreement, we believe that a material balance for radon decay products using a steady-state, well-mixed model should give reasonable predictions of working level. When incoming air is essentially free of radon or its decay products, as was found for our experiments, the following equation is obtained:(1)

$$WL = \frac{S}{(\lambda_r + I)(\lambda_a + I + R)} \left[k_1 + \frac{k_2 \lambda_a}{\lambda_b + I + R} \left[1 + \frac{\lambda_b}{\lambda_c + I + R} \right] \right] \qquad (1)$$

where WL = potential α-energy concentration of short-lived radon decay products in air in units of working level

S = radon exhalation rate per room volume in Bq/(hr·m^3)

I = air infiltration rate in 1/hr

λ = radioactive decay constant (λ_r = 0.00758/hr, λ_a = 13.7/hr, λ_b = 1.55/hr, and λ_c = 2.11/hr)

k_1, k_2 = dimensional conversion factors (3.79×10^{-4} and 2.13×10^{-4}, respectively)

R = removal rate for particulate matter, 1/hr.

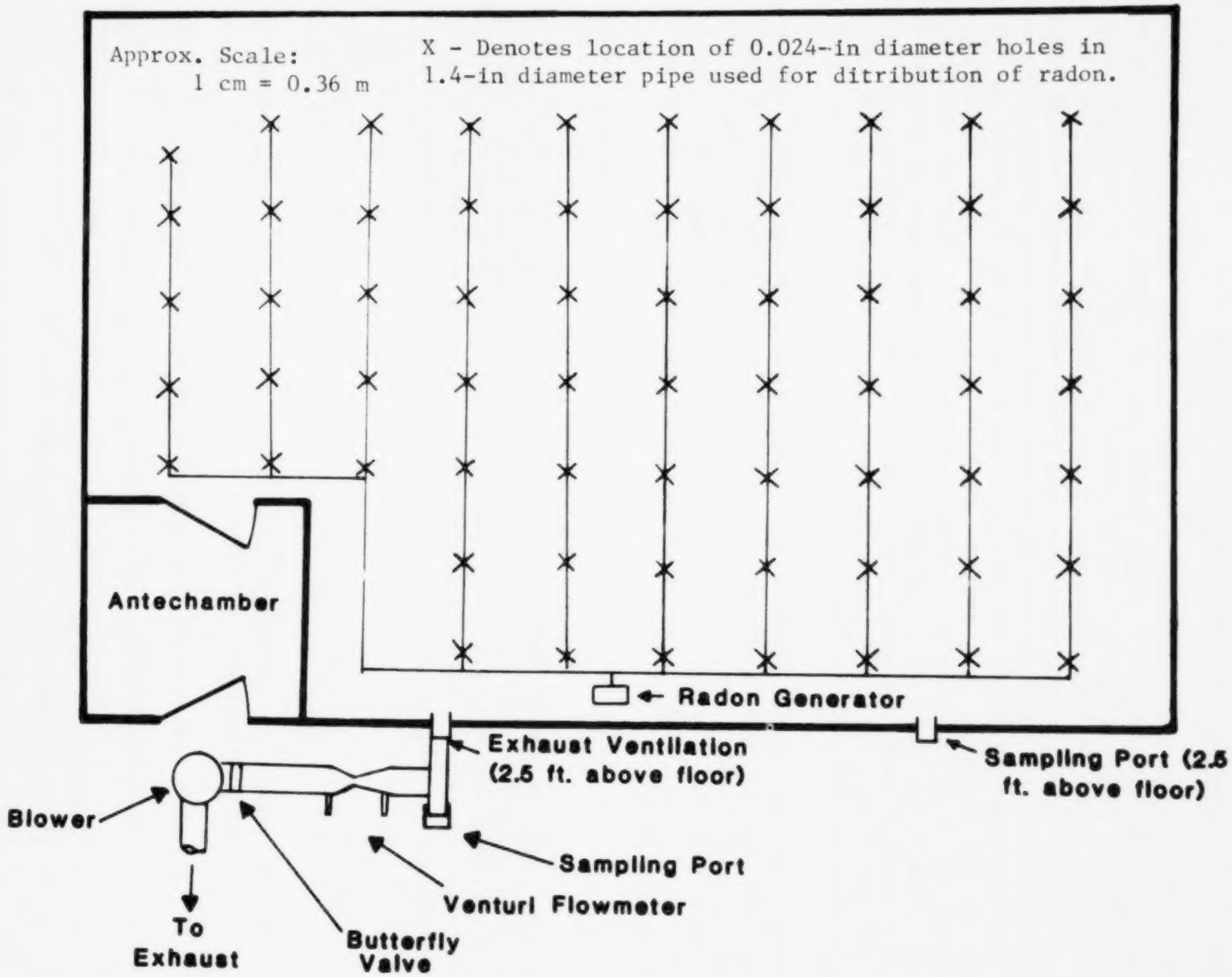
Approx. Scale:
1 cm = 0.36 m
X - Denotes location of 0.024-in diameter holes in 1.4-in diameter pipe used for ditribution of radon.
Antechamber
Radon Generator
Exhaust Ventilation (2.5 ft. above floor)
Sampling Port (2.5 ft. above floor)
Blower
Sampling Port
Venturi Flowmeter
To Exhaust
Butterfly Valve

FIGURE 1: SCHEMATIC DRAWING OF EXPERIMENTAL CHAMBER

R can be subdivided into various particulate removal mechanisms, i.e.,

$$R = P_n + P_f + A \tag{2}$$

where P_n = rate of plateout due to normal air motion (including that caused by air infiltration)

P_f = rate of plateout due to forced convection (e.g., from mixing fans)

A = air cleaning rate (e.g., from filters and electrostatic precipitators)

These terms are not necessarily independent. For example, a filter may remove particles, while its fan enhances plateout.

RESULTS

Experimental protocol was the same for all runs. The air infiltration rate was set, and, after the contents of the chamber were allowed to come to steady-state conditions, various measurements were made, some of which will be reported at a later time.(1) Whenever possible, the data obtained were compared to predicted concentrations from Equations (1) and (2).

Effect of Air Infiltration Rate

A log-log plot of the experimentally measured working level versus air infiltration rate is shown in Figure 2. (The solid line is the linear regression line.) Also shown are predicted values from Equation (1) when only plateout due to normal air motion is considered. The data follow the general slope of these lines, and a plateout rate of between 0.25 and 0.50/hr can be inferred. (A background plateout rate of 0.4/hr was assumed when the model was used for prediction in other sections of this paper.) Verification of these results by direct measurements of potential alpha energy on the surfaces of the chamber were made and will be discussed later.

Effect of Forced Convection

The effect of various mixing fans is shown in Figure 3, a plot of working level fraction remaining after a fan was started versus air infiltration rate. Reductions in WL varied from about 40 to 80%. Based on Equations (1) and (2), predicted values of WL fraction remaining versus air infiltration rate, with rate of plateout due to forced convection as a parameter, are also shown in Figure 3. From this plot, a rate of plateout due to the fans of between about 2 and 5/hr can be inferred. (This plateout rate is in addition to that obtained from normal air motion.) The data also show a greater reduction in WL at lower air infiltration rate, although the reason for this is not clear.

Manufacturer's specifications for these fans are given in Table I.

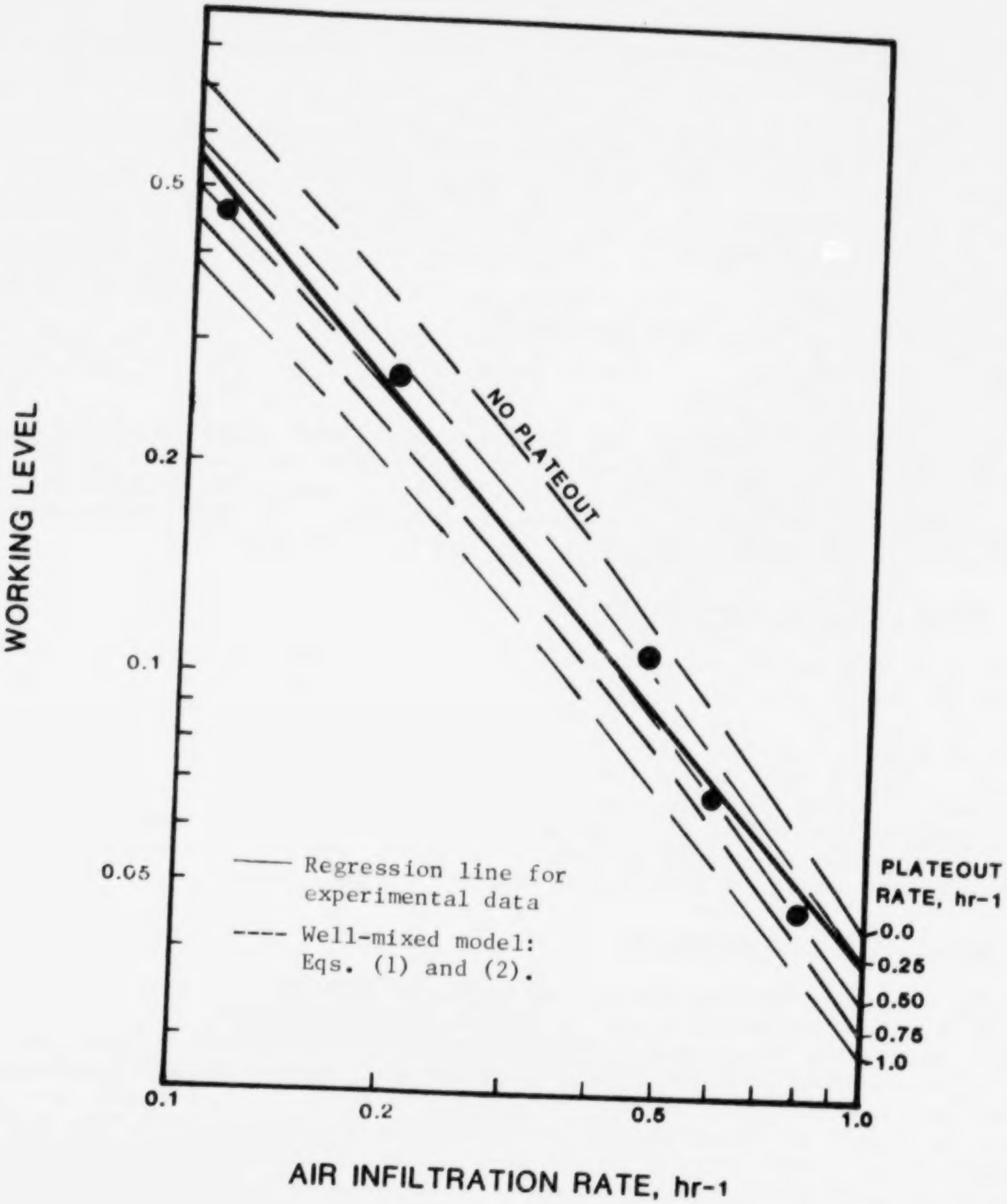

FIGURE 2: WORKING LEVEL; AIR INFILTRATION RATE WITH NO AIR TREATMENT.

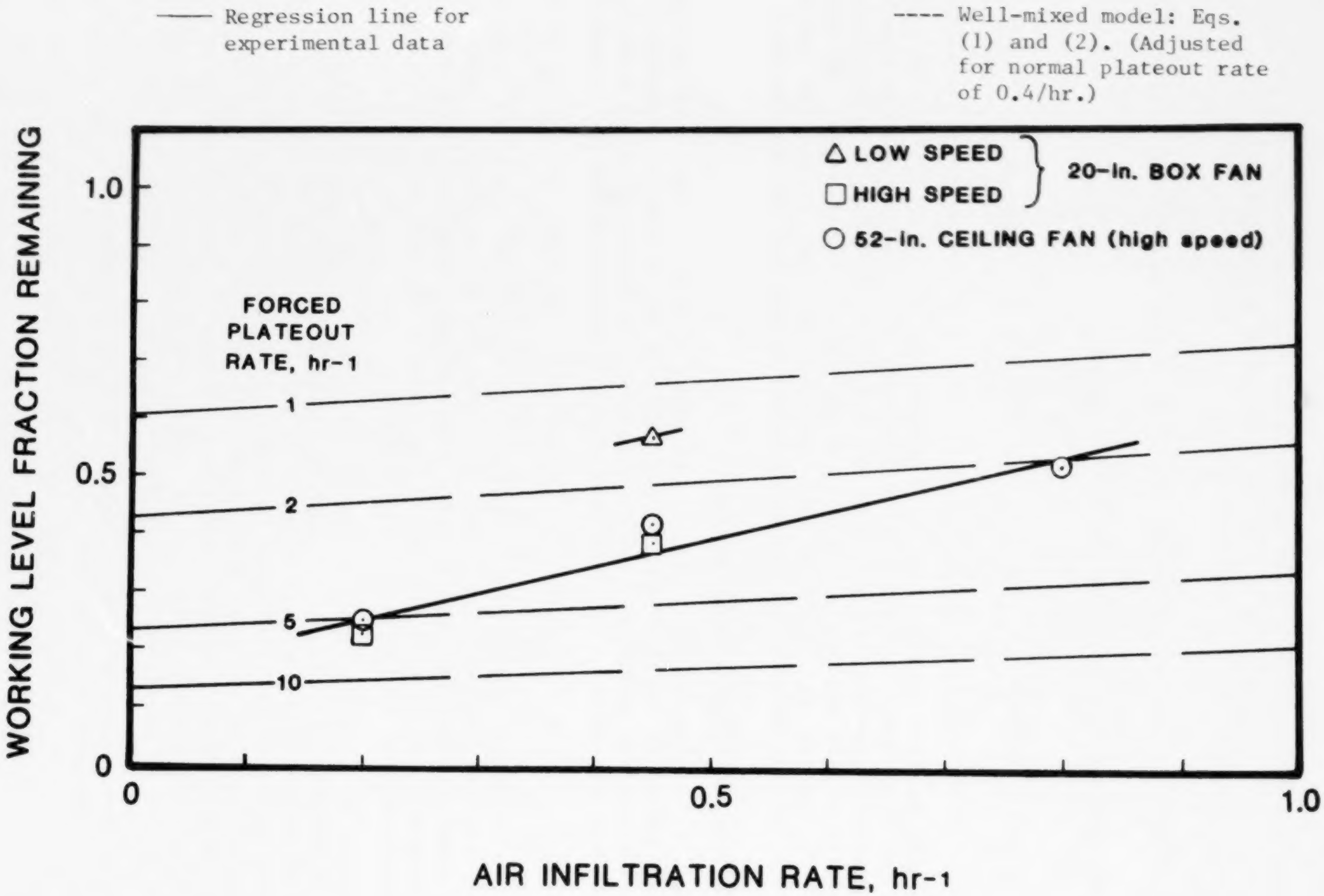

FIGURE 3: EFFECT OF MIXING FANS ON WORKING LEVEL

TABLE I:

MANUFACTURER'S SPECIFICATIONS FOR MIXING FANS				
	Speed	cfm (NEMA Method)	RPM	Watts
Hunter 20-in box fan (Model 11077)	High	5500	1000	185
	Medium	5200	930	150
	Low	4800	750	100
Hunter 52-in ceiling fan (Model 22306-7J)	High	7000	200	155
	Low	4000	115	80

Although the box and ceiling fans gave comparable reductions in WL, the ceiling fan has significant advantages: it consumes less energy than the box fan, is considerably quieter, is designed to operate continuously, and produces less noticeable air motion. It also can be used all year round; the manufacturer(10) claims it "reduces air conditioning cost during the cooling season and reduces wasteful heat stratification during the heating season."

Effect of Particulate Air Cleaners

Figure 4, a plot of WL fraction remaining versus air infiltration rate, shows the effect of various particulate-removing air cleaners. Predicted WL fractions remaining versus air infiltration rate with air cleaning rate as a parameter are also shown. (A background plateout rate of 0.4/hr was assumed.)

The electrostatic precipitators (Sears Model No. 156.73300 Console Air Cleaner), which we measured to give an air throughput rate of 5/hr for our chamber, yielded WL reductions that were in excellent agreement with the predicted values, although they assume 100% collection efficiency. About half of this reduction was obtained with the high voltage turned off. Whether this latter reduction was due to removal by the unit's fan and charcoal filter or by enhanced plateout on the walls is not known.

A high-efficiency particulate air (HEPA) filter, which removes essentially all particulate matter, was also tested at an air throughput rate of 5/hr. It yielded an effect equivalent to between 10 and 20/hr. This difference can be attributed to increased plateout. Removal of particles from the air would increase the fraction of radon decay products that are unattached to particles and thus significantly enhance plateout; air motion caused by the HEPA's blower would also promote plateout. We noted other indications of this phenomenon: limited data on the unattached fraction(1) indicated that about 80% of the working level was unattached, whereas only about 20% was unattached for most other runs. Extreme disequilibrium, another indication of high unattached fraction, was also observed.(1)

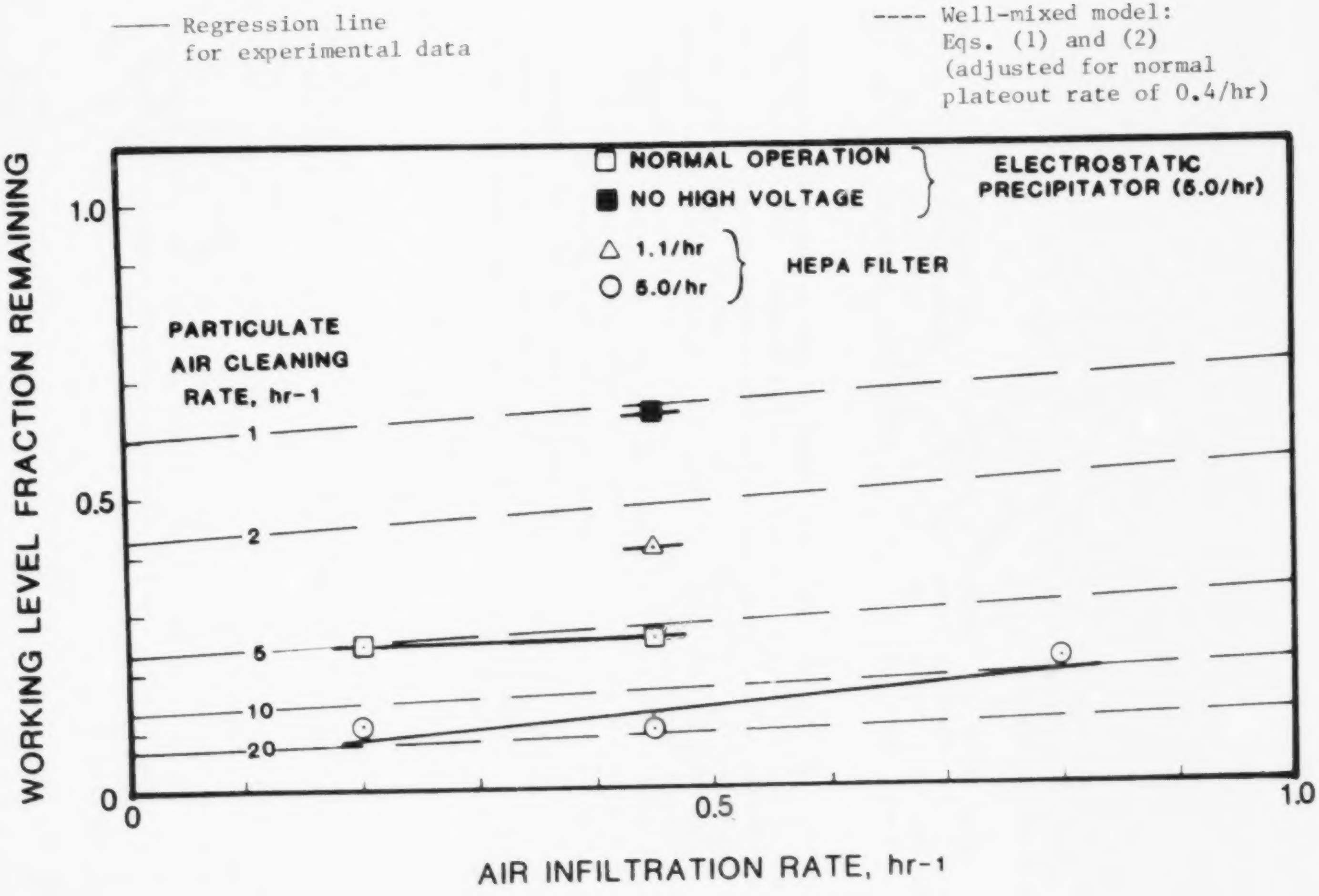

FIGURE 4: EFFECT OF PARTICULATE AIR CLEANERS ON WORKING LEVEL

DIRECT PLATEOUT MEASUREMENTS

Paper or aluminum-foil disks, affixed to various locations on the surfaces of the chamber, were removed from the chamber and counted sequentially by alpha spectrometry. The fan blades were also removed from the chamber and selected areas counted simultaneously by direct contact with six ZnS alpha scintillation counters, which were protected from room light by thin aluminized mylar windows. Counting was done over two time periods for alpha spectrometry and three time periods for scintillation, and the potential alpha-energy areal density (MeV/m^2) was back-calculated to the time at which the samples were removed from the chamber.(1)

The results of a material balance on potential alpha-energy for five runs are given in Table II. Contributions from the air, chamber surfaces, and fan blades and housing were summed and compared to the predicted values from Equation (1); between 69% and 97% of the predicted potential energy was accounted for.

Greater than 95% of the plateout was found to take place on the chamber surfaces. Holub et al.(8) found deposition on fan blades, but none on walls for similar experiments. The reasons for these conflicting results may be due to differences in measuring technique. In our experiments, less than 5% of the potential alpha-energy lost from the air was found on the fan blades, which had only 0.4% of the surface area of the chamber. Thus, the potential alpha-energy areal density on the fan blades was greater than 10 times that on the walls. This might explain why the relative external activity on the fan blades measured by Holub et al.'s beta-gamma detection system was in a measurable range, whereas the wall activity was not detectable by their equipment. Other differences between Holub et al.'s and our experiment, such as aerosol size and concentration, chamber surface to volume ratio, and infiltration air sources, make comparison difficult.

CONCLUSIONS

Various methods may be used to reduce working levels in residences. A ceiling fan, for example, was shown to give up to 75% reduction in working level. Because it was most effective at low air infiltration rates, it is probably a good choice for an energy-efficient house. It consumes about 150 W of power and may also reduce cooling and heating requirements. Greater reductions are possible using HEPA filters, but higher energy consumption and noise may prove to be a deterrent to their use. Higher fractions of unattached radon decay products found when HEPA filters were used may be less desirable from a health-effects standpoint, as the increased hazard commonly believed to be associated with unattached radon decay product may offset some of the benefits of lower working levels.

ACKNOWLEDGEMENTS

This work was supported by the U.S. Environmental Protection Agency Contract No. 68-01-6250.

TABLE II

In-Chamber Air Treatment	AIR Infiltration Rate, Hr^{-1}	MATERIAL BALANCE ON POTENTIAL ALPHA ENERGIES IN MeV[b]					
		Predicted[a] Potential α-Energy (Total)	EXPERIMENTAL MEASUREMENTS				
			AIR	CHAMBER SURFACES	FAN BLADES	FAN BOX	Found / Predicted
None	0.20	3.97×10^{9} (100%)	2.972×10^{9} (74.9%)	5.043×10^{8} (12.7%)	8.109×10^{6} (.02%)[c]	---	87.8%
None	0.45	1.52×10^{9} (100%)	1.289×10^{9} (84.8%)	1.802×10^{8} (11.8%)	9.36×10^{5} (0.1%)[c]	---	96.7%
132-cm ceiling fan (high speed)	0.20	3.97×10^{9} (100%)	8.284×10^{8} (20.9%)	2.641×10^{9} (66.6%)	1.006×10^{8} (2.5%)	---	89.9%
132-cm ceiling fan (high speed)	0.45	1.52×10^{9} (100%)	5.768×10^{8} (37.9%)	7.570×10^{8} (49.8%)	3.861×10^{7} (2.5%)	---	90.2%
Two 51-cm box fans (high speed)	0.45	1.52×10^{9} (100%)	4.56×10^{8} (30%)	5.265×10^{8} (34.6%)	4.095×10^{7} (2.7%)	2.808×10^{7} (2%)	69.3%

[a] Predicted potential alpha energy based on a well-mixed model, de-emanation of 100-μCi Ra-226 into a 90-m^3 chamber, no plateout, and radon-free infiltration air.

[b] % of the predicted total potential alpha energy is shown in parentheses.

[c] On still blades of 132-cm ceiling fan.

References

1. Rudnick, S.N., Hinds, W.C., Leith, D., First, M.W., Maher, E., and Price, J., "Effect of Indoor Air Circulation Systems on Radon Decay Product Concentration," Final Report under EPA Contract 68-01-6250 (in preparation).

2. Goodwin, A., "Review of Problems and Techniques for Removal of Radon and Radon Daughter Products from Mine Atmospheres," in Proceedings of the 12th AEC Air Cleaning Conference, CONF-720823, NTIS, Springfield, VA, 1973, pp. 378-398.

3. Coleman, R.D., Kuznetz, H.L., Woolrich, P.F., and Holaday, D.A., "Radon and Radon Daughter Hazards in Mine Atmospheres," Ind. Hyg. Quart. 17:405 (1956).

4. Shreve, J.D., and Cleveland, J.E., "Effects of Depressing Attachment Ration of Radon Daughters in Uranium Mine Atomosphere," Am. Ind. Hyg. Assoc. J. 33:304 (1972).

5. Rock, R.L., "Control of Radon Daughters in U.S. Underground Uranium Mines," in Proceedings of the 12th AEC Air Cleaning Conference, CONF-720823, NTIS, Springfield, VA, 1973, pp. 348-354.

6. Washington, R.A., Chi, W., and Regan, R., "The Use of Vermiculite to Control Dust and Radon Daughters in Underground Uranium Mine Air," in Proceedings of the 12th AEC Air Cleaning Conference, NTIS, Springfield, VA, 1973, pp. 355-376.

7. Wrenn, M.E., Eisenbud, M., Costa-Ribeiro, C., Hazle, A.J., and Siek, R.D., "Reduction of Radon Daughter Concentrations in Mines by Rapid Mixing Without Makeup Air," Health Phys. 17:405 (1969).

8. Holub, R.F., Droullard, R.F., Ho, W., Hopke, P.K., Parsley R., and Stukel, J.J., "The Reduction of Airborne Radon Daughter Concentration by Plateout on an Air Mixing Fan," Health Phys. 36:497 (1979).

9. Thomas, J.W., "Measurement of Radon Daughters in Air," Health Phys. 23:783 (1972).

10. Robins & Myers, Inc., manufacterer's literature provided to retailers.

SOME MEASUREMENTS OF THE EQUILIBRIUM FACTOR FOR RADON DAUGHTERS IN HOUSES*

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ABSTRACT

The problem of relating radon levels in houses to the radiation doses received by their residents would be greatly simplified if the parameters which determine the equilibrium factor, or working level ratio, could be identified and their effects predicted. The first step in such a program is to measure the equilibrium factor in a number of houses under a variety of conditions, by simultaneously measuring both the radon concentration and the working level.

Our preliminary measurements in a few houses indicate that the equilibrium factor normally varies over a range of 0.05 to 0.50, but on occasions it approaches 1.0. The mean of eight sets of observations was 0.20 ± 0.14 (one std. dev.). Some of the parameters which have been observed to affect the equilibrium factor include the relative areas of room surfaces and airborne particles, the ventilation rate, and the air circulation. Human activities, such as cooking and smoking, also affect the equilibrium factor, by directly affecting the aforementioned variables.

INTRODUCTION

Because the radiation dose to the lung from environmental radon is due almost entirely to the inhalation of the short-lived daughter products, measurements of radon levels alone are not sufficient to determine the dose. Typically the activities of the daughter products in air are not in equilibrium with the parent radon, nor is there equilibrium among the daughters themselves. The working level (WL) was introduced as a convenient unit to overcome this problem in uranium mines and it is also applicable to other environments where radiation dosimetry is the principal concern. It is defined as that concentration of short-lived radon daughters in any combination which has a potential alpha-particle energy of 1.3×10^5 MeV per liter of air. Since

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*Work supported by U.S. Department of Energy under contract #W-31-109-ENG-38.

+Visiting Scientist from the North China Institute of Radiation Protection, Taiyuan, Shanxi, PRC.

this is equal to the potential alpha-particle energy of 100 pCi/l of radon with all its short-lived daughters in equilibrium, we can define the equilibrium factor, F, as one hundred times the working level divided by the radon concentration in pCi/l. Since it is easier to determine the radon concentration than the working level, the task of determining lung doses would be simplified if those factors which determine F could be identified and their effects quantified. In order to do this, simultaneous measurements of radon concentrations and working levels must be made in houses under a variety of conditions. In addition, parameters which may affect the equilibrium factor, such as aerosol concentration and ventilation rate, must also be identified and monitored. Although the effects of such parameters may be best determined under controlled conditions in a laboratory, the results must be validated by measurements in houses.

MEASUREMENTS OF F

Radon levels were determined by either collecting air samples in Lucas flasks (Lu57), or by continuous monitoring with a Spitz-Wrenn chamber (Wr75). Concentrations of radon daughters were determined and working levels computed with an Environmental Working Level Monitor (Ke78). Measurements were made in three houses owned and occupied by ANL employees; two of the three (H-01, H-23) have radon levels which routinely exceed 370 Bq/m^3 (10 pCi/l). The radon levels in the third (H-02) ranged from 11 to 300 Bq/m^3 (0.3 - 8.0 pCi/l). The results are shown in Table 1.

Values for F reported by other workers have averaged around 0.5 (Ge78, McG80). Only one of our series of observations approached this value, while the mean of all our observations is 0.20 ± 0.14 (one std. dev.). The radon concentration (Spitz-Wrenn chamber), working level (EWLM), and equilibrium factor in the basement of house 23 during the period 27-30 Sep are shown in Fig. 1. Note that while the radon concentration and working level undergo diurnal variations by a factor of four, 88% (71/81) of the values of F lie within the range 0.15 - 0.30. However, some of the variability in F may be an artifact of the measurement techniques. The EWLM determines working level on the basis of a three-minute sample of air, whereas the values obtained with the Spitz-Wrenn chamber were averages of 60-minute counts; furthermore, there is a delay between a change in the radon concentration outside the chamber and equilibration inside it, and another delay before the electrostatically collected daughter products are at equilibrium with the radon producing them.

Although the variations in values of F have not as yet been firmly linked to such parameters as air exchange rate or airborne particle concentrations, we have observed the lowest values of F when the fan of the heating or cooling system is in operation. Presumably this removes dust particles from the air by filtration and impaction, giving unattached daughter products the chance to be removed from the air by deposition on surfaces within the house.

MEASUREMENTS OF SURFACE DEPOSITION

Laboratory experiments have shown that radon daughters "plate out," i.e., deposit on various surfaces exposed to a radon-containing atmosphere (Ho79, Ge81). We have recently performed some direct measurements of radon daughter plate out on surfaces in a house. The data are shown in Fig. 2. When the cover was removed from the window (316 cm^2) of a gas-flow proportional counter, the counting rate immediately increased, presumably due to the detection of the decay of radon and daughters in the air within α-particle range of the counter (about 4 cm). The counting rate then continued to increase, showing the characteristic build-up of activity due to deposition of radon daughters on the surface of the window, until an equilibrium was reached. When the cover was replaced, the counting rate doubled, because radon daughters were also deposited on the surface of the cover itself, and then decreased at a rate characteristic of a mixture of radon daughters.

SUMMARY

Simultaneous measurements of radon levels and working levels in a few houses have resulted in a mean value for the equilibrium factor of 0.20 ± 0.14. The lowest values of F were observed during the operation of the fan of a house's heating or cooling system, which presumably removed radon daughters from the air by both filtration and impaction on walls and other surfaces. Direct observations of the plate out of radon daughters on surfaces in a house were performed, confirming the results of laboratory experiments by other workers.

References

Ge80 A.C. George and A.J. Breslin, 1980, The distribution of ambient radon and radon daughters in residential buildings in the New Jersey-New York area, in Natural Radiation Environment III, T.F. Gesell and W.M. Lowder, Eds., U.S. Department of Energy Symposium Series No. 51, CONF-780422, Vol. 2, p. 1272.

Ge81 A.C. George and E.O. Knutson, 1981, Measurements of radon daughter plateout, presented at the 26th Annual Meeting of the Health Physics Society, Louisville, KY, June 21-25, 1981, Abstract No. P/189.

Ho79 R.F. Holub, R.F. Droullard, W. Ho, P.K. Hopke, R. Parde, and J.J. Stukel, 1979, The reduction of airborne radon daughter concentration by plateout on an air mixing fan, Health Phys. 36, 497.

Ke78 D.J. Kefe, W.P. McDowell, an P.G. Groer, 1978, The environmental working level monitor, Final Report to U.S. Environmental Protection Agency, Argonne National Laboratory Report #P7628C.

Lu57 H.F. Lucas, Jr., 1957, Improved low-level alpha-scintillation counter for radon, Rev. Sci. Inst. 28, 680.

McG80 R.G. McGregor, P. Vasudew, E.G. Letourneau, R.S. McCullough, F.A. Prantl, and H. Taniguchi, 1980, Background concentrations of radon and radon daughters in Canadian homes, Health Phys. 39, 285.

Wr75 M.E. Wrenn, H. Spitz, and N. Cohen, 1975, Design of a continuous digital-output environmental radon monitor, IEEE Trans. Nucl. Sci. NS-22, 645.

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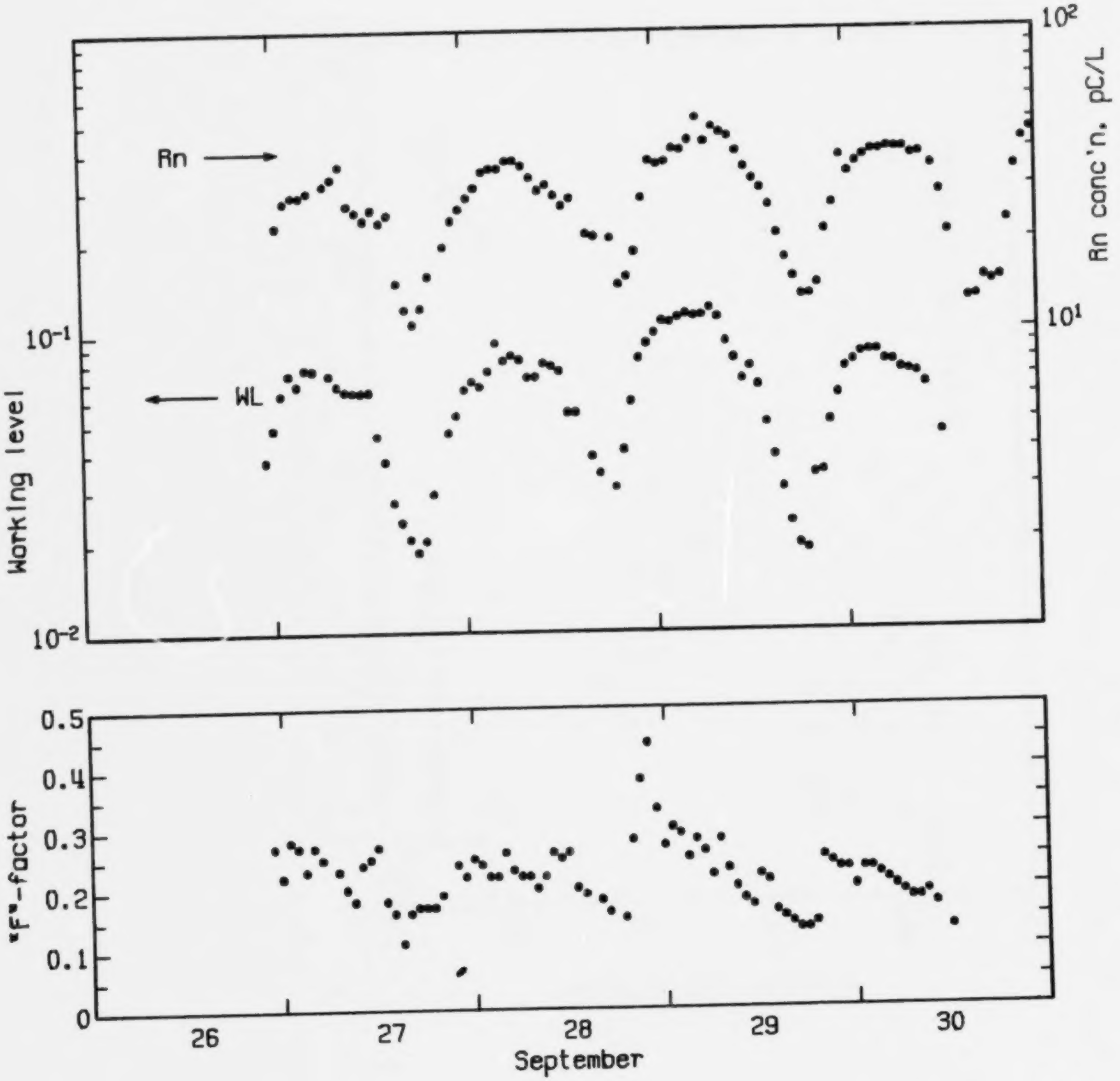
Table 1. Observed values for the equilibrium factor, F.

House	Location	Period	No. Observations	Mean F	Range of F
H-01	2nd floor	28-31 Jul	7	0.48 ± 0.21	0.19 - 0.83
H-01	2nd floor	25-30 Aug	9	0.16 ± 0.11	0.02 - 0.35
H-01	2nd floor	15-16 Feb	28	0.07 ± 0.04	0.03 - 0.20
H-01	2nd floor	1-6 Mar	93	0.09 ± 0.03	0.04 - 0.23
H-02	1st floor	17-21 Aug	11	0.31 ± 0.13	0.12 - 0.60
H-23	Basement	26-30 Sep	81	0.22 ± 0.06	0.11 - 0.44
H-23	1st floor	14-15 Oct	23	0.13 ± 0.02	0.08 - 0.16
H-23	1st floor	7-9 Jan	57	0.12 ± 0.03	0.06 - 0.21

FIGURES

Fig. 1. The radon concentration, working level, and equilibrium factor in the basement of house no. H-23 for a period of four days.

Fig. 2. The build-up and decay of radon daughters on the window of a large-area proportional counter placed in the utility room of house no. H-01.


Rn
WL
Working level
10⁻¹
10⁻²
Rn conc'n, pC/L
10²
10¹
"F"-factor
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0.4
0.3
0.2
0.1
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26
27
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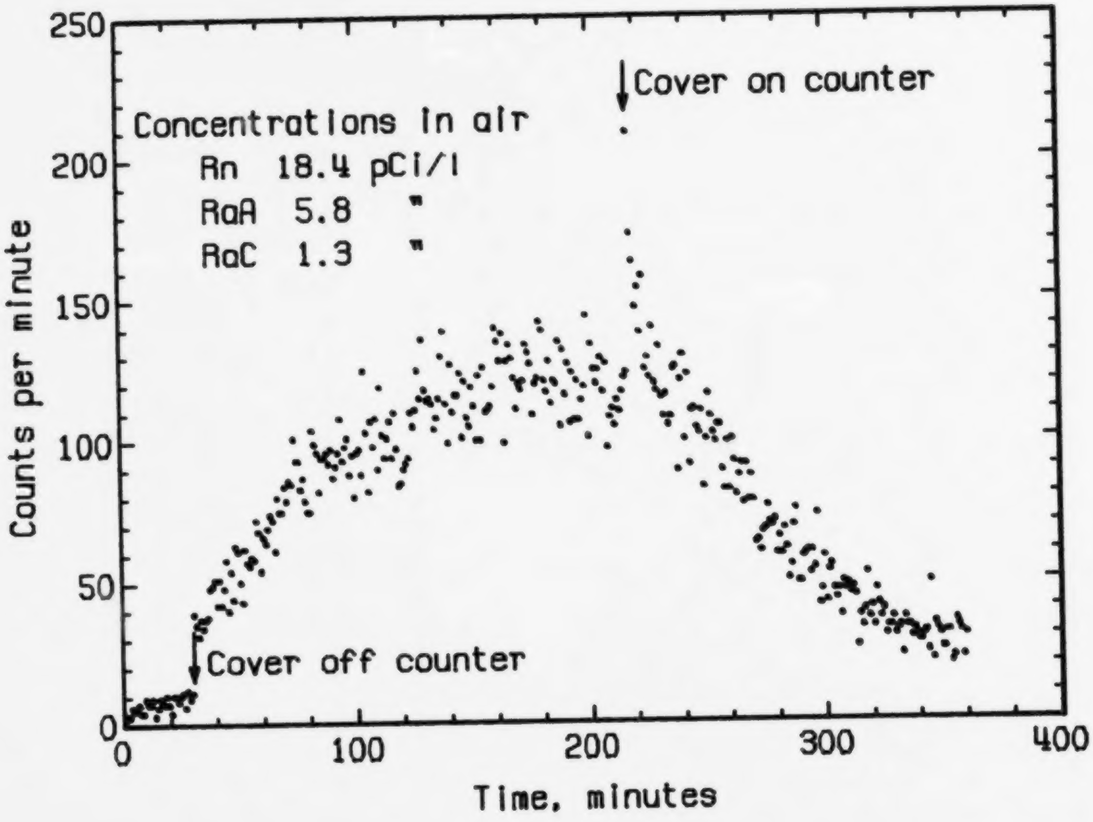
September

Figure 1

Figure 2



21 AUG 1981

REGIONAL ENVIRONMENTAL DOCUMENTATION OF NATURAL RADIATION IN SWEDEN

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ABSTRACT

In 1979, when the problem of high radon daughter levels in Swedish houses became widely publicized, the need for information on variations in the natural radiation environment became very apparent. The radon problem was at first attributed to radon emanation from alum shale based aerated concrete, but it was soon obvious that ground with an abnormally high uranium content constitutes an even greater risk for high radon and radon daughter levels in houses. The Geological Survey of Sweden was commissioned to produce a documentation in map form of all areas and rock types with gamma ray levels exceeding 30 μR/h, with the intention of delimiting risk areas for high soil gas radon contents.

The maps, known as GEO-radiation maps, are produced at the scale of 1:50 000. They are based primarily upon airborne radiometric surveys, ground measurements of gamma radiation and geological mapping. To date some 450 map sheets have been published covering approximately 55% of the country. The maps provide primary information to local planning, health and building authorities as to variations in the natural radiation environment. Within the so-called risk areas marked on the maps, local authorities are recommended to investigate the soil gas radon content prior to any new development.

Geological environments known, in Sweden, to be associated with radon daughter problems in dwellings are alum shale, a Cambrian, uranium-rich black shale, uranium-rich granites and uranium-rich pegmatites. Both alum shale and uranium-rich granites constitute extensive areas of bedrock.

INTRODUCTION

In 1978, the National Institute for Radiation Protection measured radon daughter levels in a number of houses built on uranium-rich shale tailings (Swedjemark et al. 1979). The measured levels exceeded the maximum level of 1110 Bq m^{-3} (Becquerel per cubic metre) of radon in equilibrium with radon daughters permitted in Swedish mines. This was the first proof in Sweden that radon emanating from the ground can give rise to such high concentrations of radon daughters in dwellings. Earlier, the radon problem in houses was attributed to radon emanation from building materials containing unusually large amounts of radium, for example, alum shale based aerated concrete (Swedjemark 1978a, 1979b, 1980).

Early in 1979, a Government Commission, termed the Radon Commission, was set up to investigate the problem of radiation risks in dwellings (Radonutredningen, 1979). At an early stage in the Commissions work, it became apparent that radon emanating from the ground could constitute as great, if not greater problem than radon emanation from building materials. Measurement programs carried out by Local Health Authorities during the last year have proved this to be true. In Sweden, areas with particular risk for high soil gas radon contents are considered to be those with a bedrock of uranium-rich alum shale (a Cambrian, black shale), uranium-rich granite or uranium-rich pegmatites and areas of glacial drift cover comprised largely of these rock types. Knowledge of the geographical distribution of these areas became thus necessary in order, at least, to prevent future building without the use of necessary technical measures against radon penetration from the ground.

NATURAL RADIATION ENVIRONMENT

The source of natural radiation from the ground is almost entirely related to the presence in the bedrock or drift cover of the radioactive elements uranium and thorium, and their daughter products including radon, and potassium-40. Their concentrations in various rock types in Sweden is very variable. (Table 1).

The geographical distribution of the uranium-rich alum shale formation (Fig. 1) is fairly well-known due to mapping and prospecting activities, the latter owing to the rocks' unusually high contents of uranium, vanadium, molybdenum and organic matter. The uranium content can range from 50 - 350 ppm (Armands 1972: Andersson et al. in press).

Uranium- and thorium-rich granites and pegmatites are known from a number of areas in Sweden (Fig. 2). The granites (and pegmatites) occur in a variety of geological settings in the Swedish Precambrian, and represent a variety of granite types and ages, 1750-890 Ma (Wilson and Akerblom 1980). Knowledge of the occurrence and extent of the granites and pegmatities has been obtained mainly from airborne radiometric surveys and uranium prospecting carried out principally by the Geological Survey of Sweden (SGU).

For the purpose of documenting the natural radiation environment in Sweden, and for delimiting risk areas for high soil gas radon contents, the Geological Survey of Sweden was commissioned to produce a documentation in map form of all areas and rock types known to be particularly radioactive. The maps, known as GEO-radiation maps, are produced at the scale of 1:50 000. They are based primarily upon airborne radiometric surveys, ground measurements of gamma radiation and geological mapping.

The choice of this method for delimiting risk areas for high soil gas radon contents was determined by:

	U ppm	Th ppm	K %	m_{Ra}	µR/h
Granite, normal	2 - 10	5 -20	2 -6	0.1 - 0.6	5 - 20
Granite U- and Th-rich	8 - 40	10 -90	4 -6	0.5 - 2.5	12 - 65
Gneiss	2 - 10	5 -20	2 -6	0.1 - 0.6	5 - 20
Diorite	0.1- 2	1 -10	1 -3	0.01- 0.1	2 - 10
Sandstone	0.5- 5	1 -10	1 -5	0.03- 0.3	2 - 15
Limestone	0.5- 2	0.1- 2	0.1-0.5	0.03- 0.1	0.5- 3
Shale	1 - 10	2 -15	2 -6	0.06- 0.6	5 - 18
Alum Shale	10 -350	2 -10	3.5-6	3.1 -21.5	10 -230

Table 1: Uranium, thorium and potassium contents, radium index and gamma radiation for different rock types in Sweden.

Figure 1

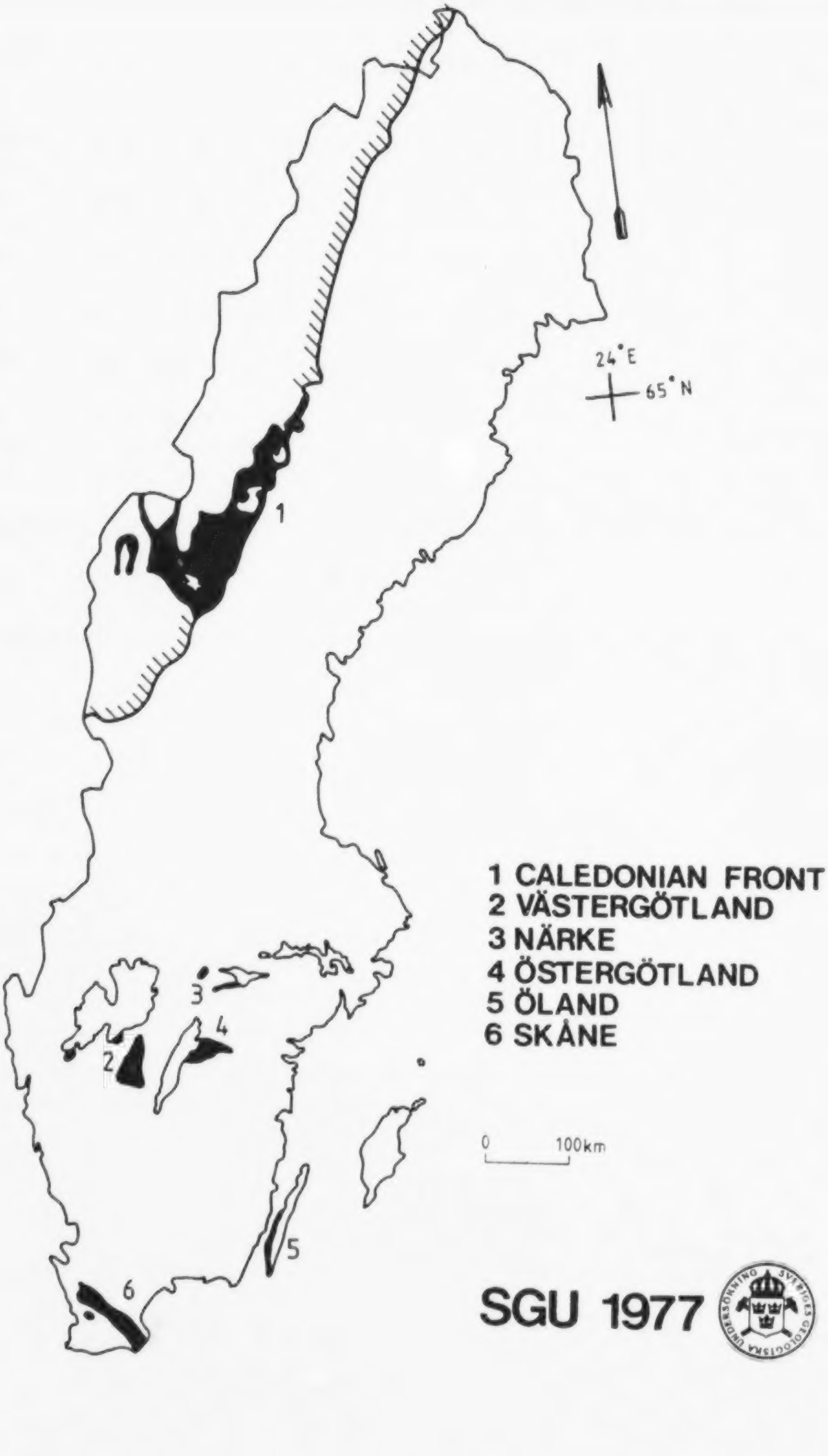
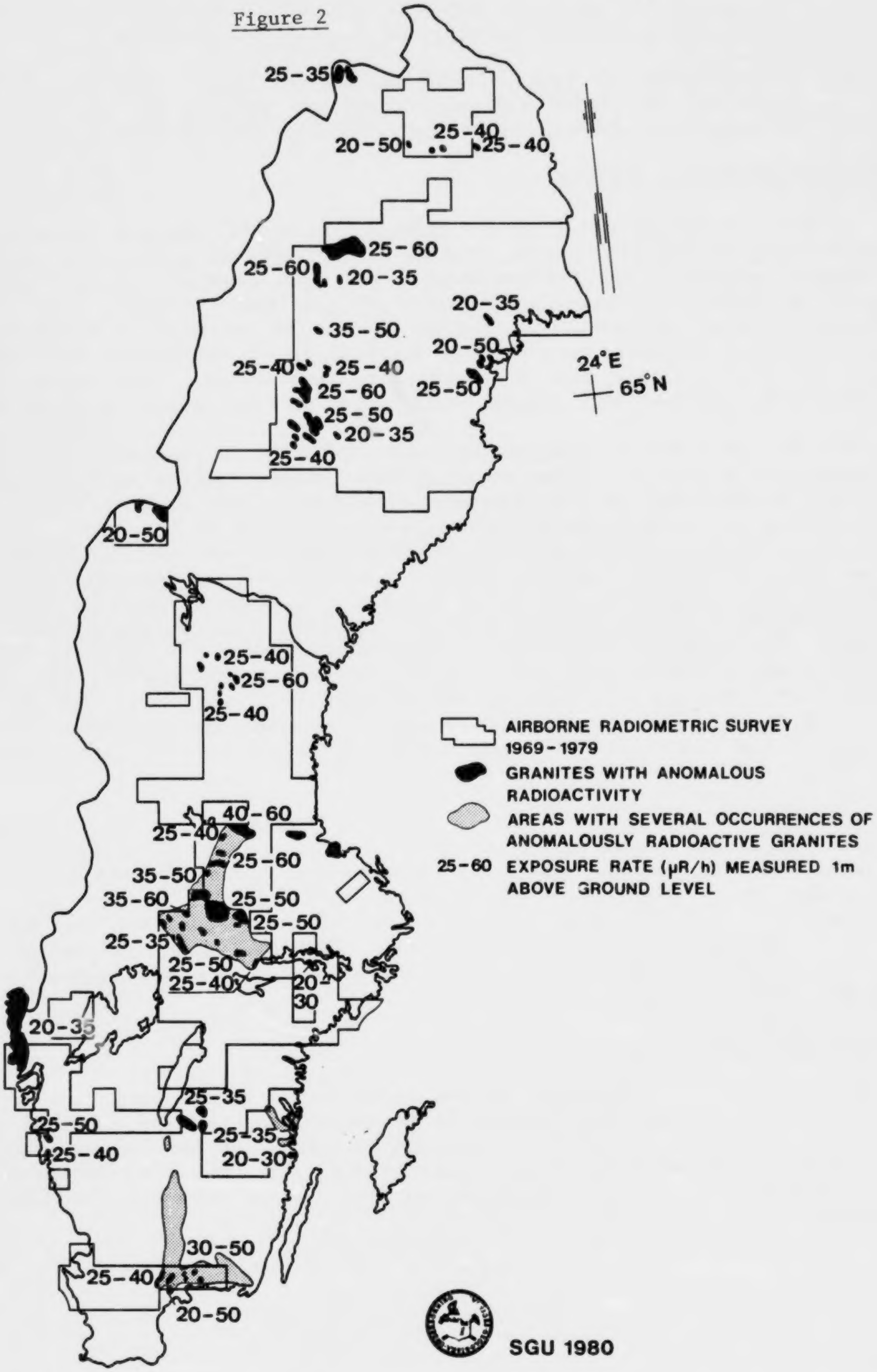

24°E
65°N
1
2
3
4
5
6
1 CALEDONIAN FRONT
2 VÄSTERGÖTLAND
3 NÄRKE
4 ÖSTERGÖTLAND
5 ÖLAND
6 SKÅNE
0
100km
SGU 1977

Figure 2



25-35
20-50
25-40
25-40
25-60
25-60
20-35
20-35
35-50
20-50
25-40
25-40
25-60
25-50
25-50
20-35
25-40
24°E
65°N
20-50
25-40
25-60
25-40
40-60
25-40
25-60
35-50
35-60
25-50
25-50
25-35
25-50
25-40
20
30
20-35
25-35
25-50
25-35
25-40
20-30
30-50
25-40
20-50
AIRBORNE RADIOMETRIC SURVEY 1969-1979
GRANITES WITH ANOMALOUS RADIOACTIVITY
AREAS WITH SEVERAL OCCURRENCES OF ANOMALOUSLY RADIOACTIVE GRANITES
25-60 EXPOSURE RATE (μR/h) MEASURED 1m ABOVE GROUND LEVEL
SGU 1980

1. the assumption that the greatest concentrations of radon in the soil are directly related to rocks and soils rich in uranium.

2. availability of regional information on gamma radiation from the airborne and ground radiometric surveys carried out by SGU. These surveys cover to date approximately 45% of the country.

AIRBORNE RADIOMETRIC SURVEYS

Airborne gamma-ray spectrometry carried out by SGU is used primarily for regional prospecting for uranium, and as a complementary geophysical method to geological mapping. The instrumentation has been designed to cope with the special problems of prospecting in areas of glaciated terraine. Most of the bedrock in Sweden is covered by glacial till which can vary in thickness from 4 to 10 or more metres. Much of this material is of relatively local derivation, but it has always been transported to some extent. This latter factor is of special interest with regard to the radon problem as will be shown later.

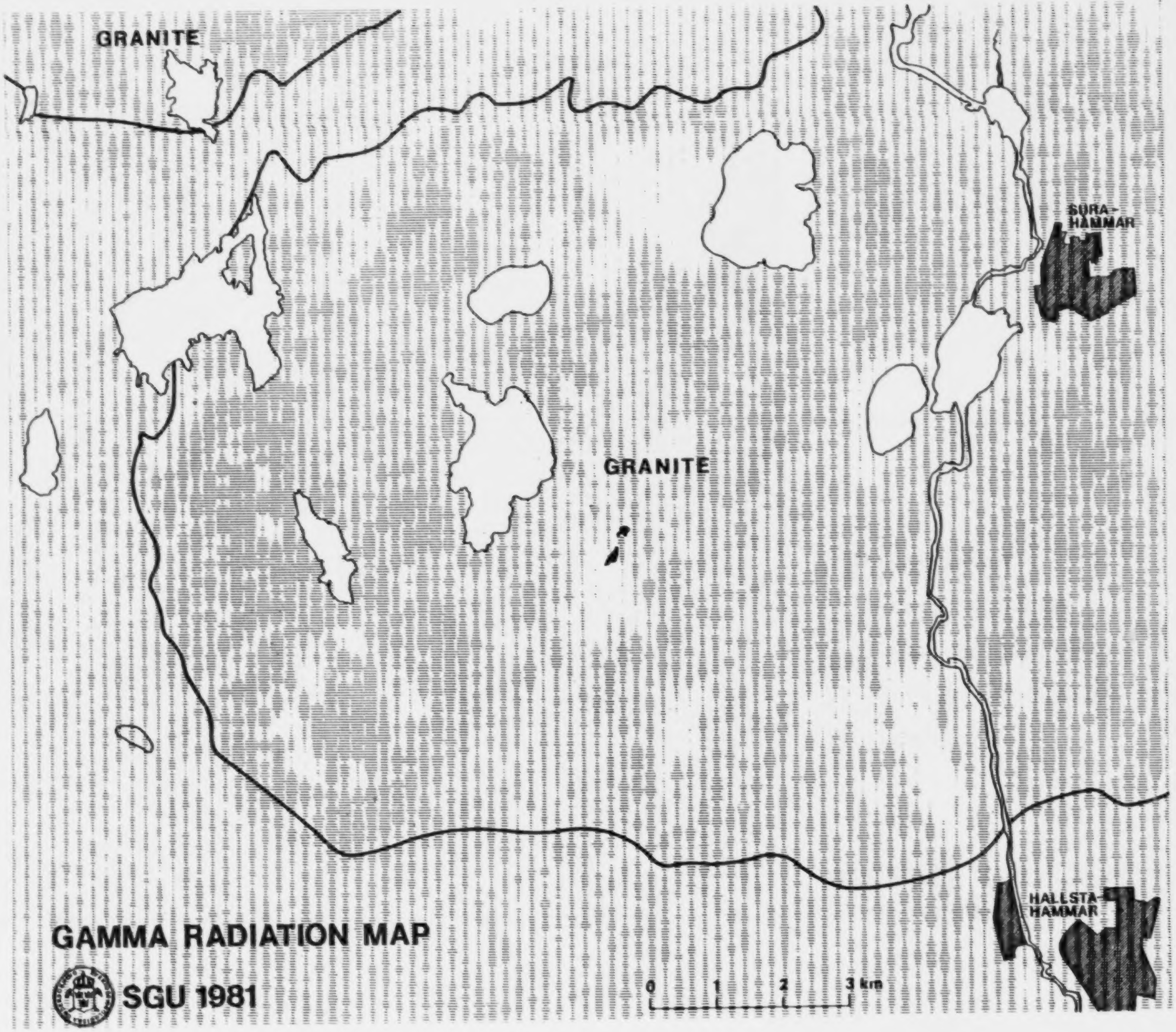
The SGU technique (Linden and Akerblom, 1976) is to use four sodium iodide crystals giving a total volume of 17 litres. Gamma spectra between 0.45 and 2.85 MeV are recorded on 258 channels. The plane flies at an altitude of 30 metres along profiles 200 metres apart, and at a speed of 70 m/sec. Registration takes place digitally with a measuring time of 400 msec, readings being taken every 40 metres. After computer processing of the data from all the channels, the results of the measurements are plotted automatically as maps using an ink-jet color plotter. Gamma radiation from uranium, thorium and potassium for each measuring station is marked on the maps as colored lines (U red, Th blue and K yellow), the lengths of which are proportional to the registered equivalent contents of these elements (U and Th as ppm and K as %). The three-component, colored, gamma radiation maps give a detailed picture of the distribution of the radioactive elements over large areas, as well as locating point anomalies (Fig. 3a and b).

Follow-up of the anomalous points or areas for each gamma radiation map is carried out on the ground using handborne scintillometers. Observations as to terraine, bedrock geology, Quaternary geology and gamma-ray levels are noted for areas where gamma-spectrometry measurements on the ground have been carried out. In the compilation of the GEO-radiation maps, all this material is used together with published geological and Quaternary geological maps (where available), and detailed local knowledge of the geologists mapping specific areas.

GEO-RADIATION MAPS

Maps showing radon contents in the ground would be the ideal for environmental purposes. Such maps cannot be produced owing to the enormous costs involved and the length of time necessary to carry out measurement programs. An alternative would be to produce maps showing the uranium content (in ppm) of the ground, but again, this is not yet feasible for more than limited areas in Sweden.

Figure 3a


GRANITE
SURA-HAMMAR
GRANITE
HALLSTA-HAMMAR
GAMMA RADIATION MAP
SGU 1981
0 1 2 3 km

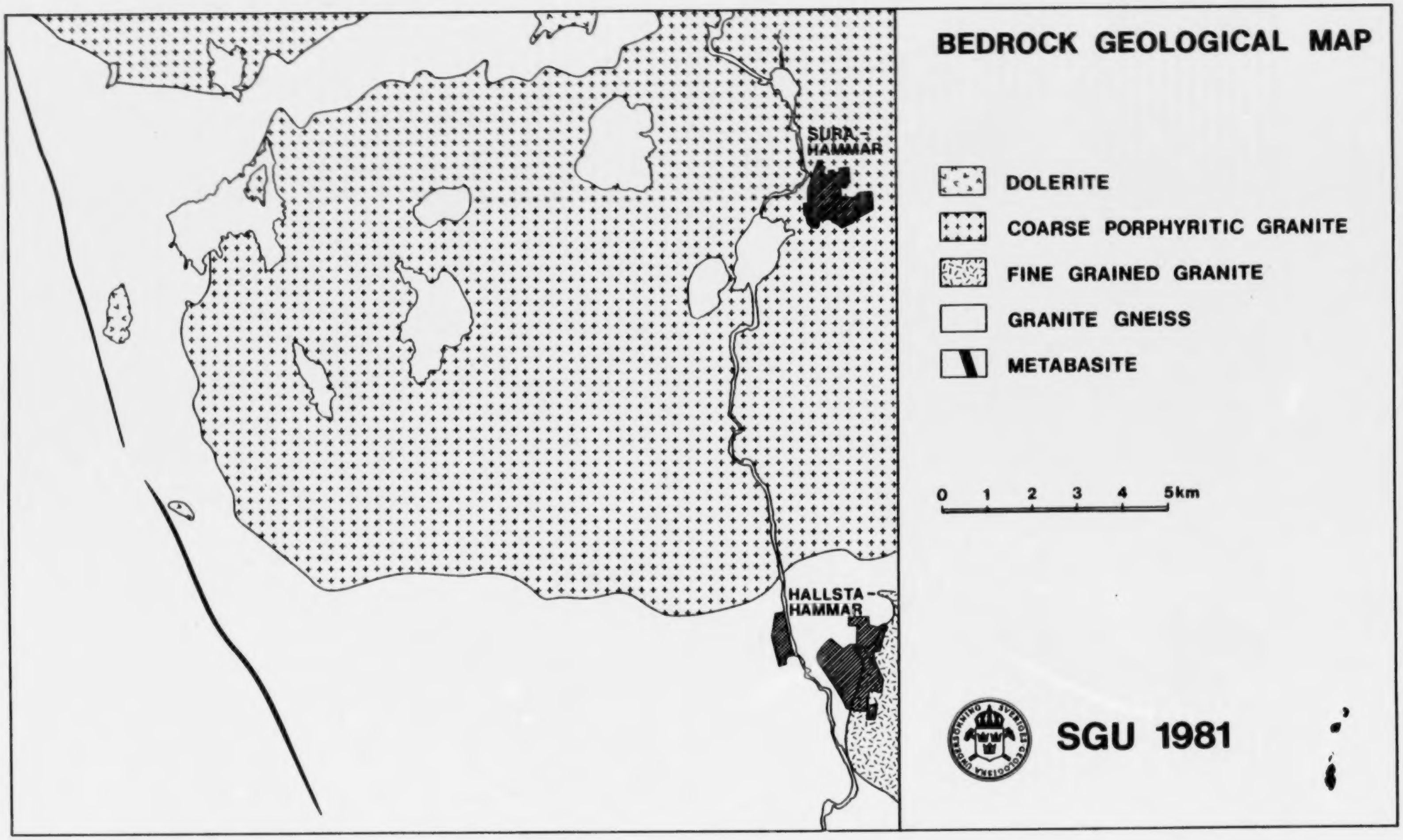

BEDROCK GEOLOGICAL MAP
DOLERITE
COARSE PORPHYRITIC GRANITE
FINE GRAINED GRANITE
GRANITE GNEISS
METABASITE
0 1 2 3 4 5km
SURA-HAMMAR
HALLSTA-HAMMAR
SGU 1981

Figure 3b

The GEO-radiation maps in their present form are intended as key maps to show areas where the level of gamma radiation from uranium and/or thorium in the bedrock or drift cover is particularly high. For this purpose the limit of 30 µR/h has been arbitrarily selected. To date some 450 maps have been produced covering approximately 55% of the country. The maps cover initially areas of alum shale and known uranium-rich granites. The latter category coincides in general with areas covered by airborne radiometric surveys. The maps are accompanied by short commentaries on the geology and radioactivity of the areas concerned. Examples of the maps will now be discussed.

Fig. 4 shows the geological map over part of the alum shale occurrence in Ostergotland, southeastern Sweden, and Fig. 5 is the GEO-radiation map over the same area. An enlarged detail from the gamma radiation map over Fornasa is shown in Fig. 6. The lines on this map are related to gamma radiation from uranium in the near surface alum shale.

The profile in Fig. 7 shows how Quaternary ice transported fragments of alum shale from the bedrock exposure up towards the surface of the till in a southerly direction. The alum shale fragments are successively mixed with other material during transport.

One can see how individual "nappes" of the shale material develop in the drift cover, the latter being about 15 metres thick. Comparison between the bedrock geological map and the gamma radiation map reveals that alum shale fragments are present in the drift cover several kilometres south of the bedrock source. In consequence, higher than normal levels of gamma radiation, and probably enhanced radon emanation, can occur beyond the area of bedrock exposure of a radioactive rock. Consideration must therefore always be given to the direction and length of transport of rock fragments when judging the risks for high soil gas radon contents.

A further illustration of this point is made in Fig. 8 which shows the gamma radiation map from an area of granite gneiss in central Sweden. The bedrock is largely concealed by a drift cover of glacial till. A clearly uranium anomalous area is distinguished south of Svennevad. This area coincides with a glacial esker and post glacial washed sands. The esker has its source some 10 kilometres to the north of Svennevad in an area of alum shale bedrock. The gamma radiation map therefore indicates spread of alum - shale fragments of the esker and related sands. Enhanced gamma radiation from uranium concentrations in the granite gneiss bedrock is otherwise only a local feature in connection with pegmatites and zones of migmatisation.

The gamma radiation, geological and GEO-radiation maps over a uranium- and thorium-rich granite in Molndal, south of Gothenburg, are shown in Figs. 9, 10 and 11. The suburb of Balltorp is planned to house 15,000 people.

The granite in question is a red to grey alkali granite which partially weathers to a coarse sand. The granite has a uranium content of 10-30 ppm, a thorium content of 30-70 ppm and radioactivity of 30-60 µR/h. The granite is bounded to the east by gneissose granodiorite and to the west by a porphyritic granite with normal uranium and thorium contents. The whole area is well exposed and the till cover, where present, is only a half to two metres thick. The valleys contain sediments, mainly clay. Glacial transport of till material was from east to west.

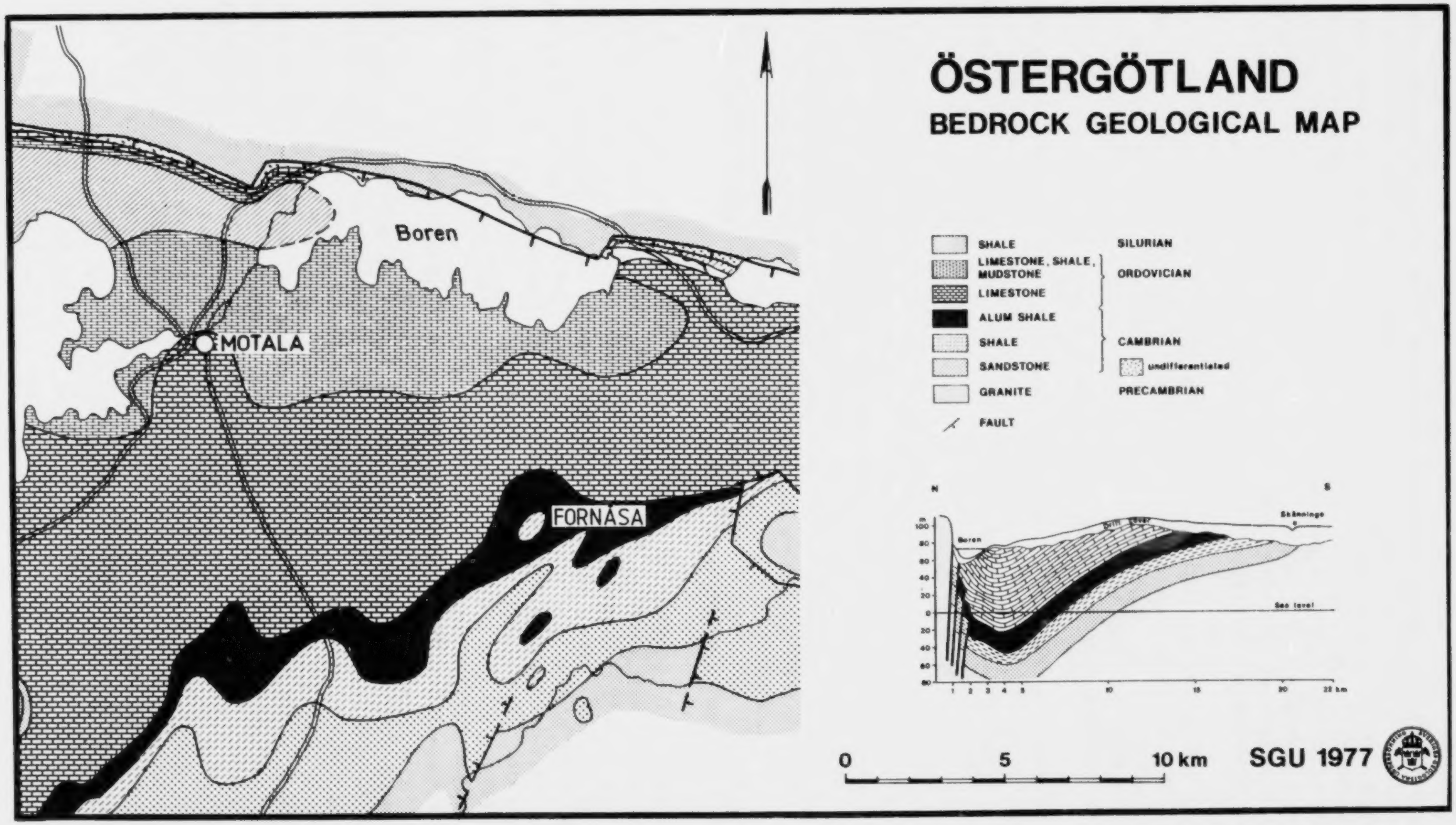

ÖSTERGÖTLAND
BEDROCK GEOLOGICAL MAP
SHALE
LIMESTONE, SHALE, MUDSTONE
LIMESTONE
ALUM SHALE
SHALE
SANDSTONE
GRANITE
FAULT
SILURIAN
ORDOVICIAN
CAMBRIAN
undifferentiated
PRECAMBRIAN
Boren
MOTALA
FORNÅSA
N
S
m
100
80
60
40
20
0
20
40
60
80
Boren
Skänninge
Sea level
1
2
3
4
5
10
15
20
22 km
0
5
10 km
SGU 1977

Figure 4

Figure 5

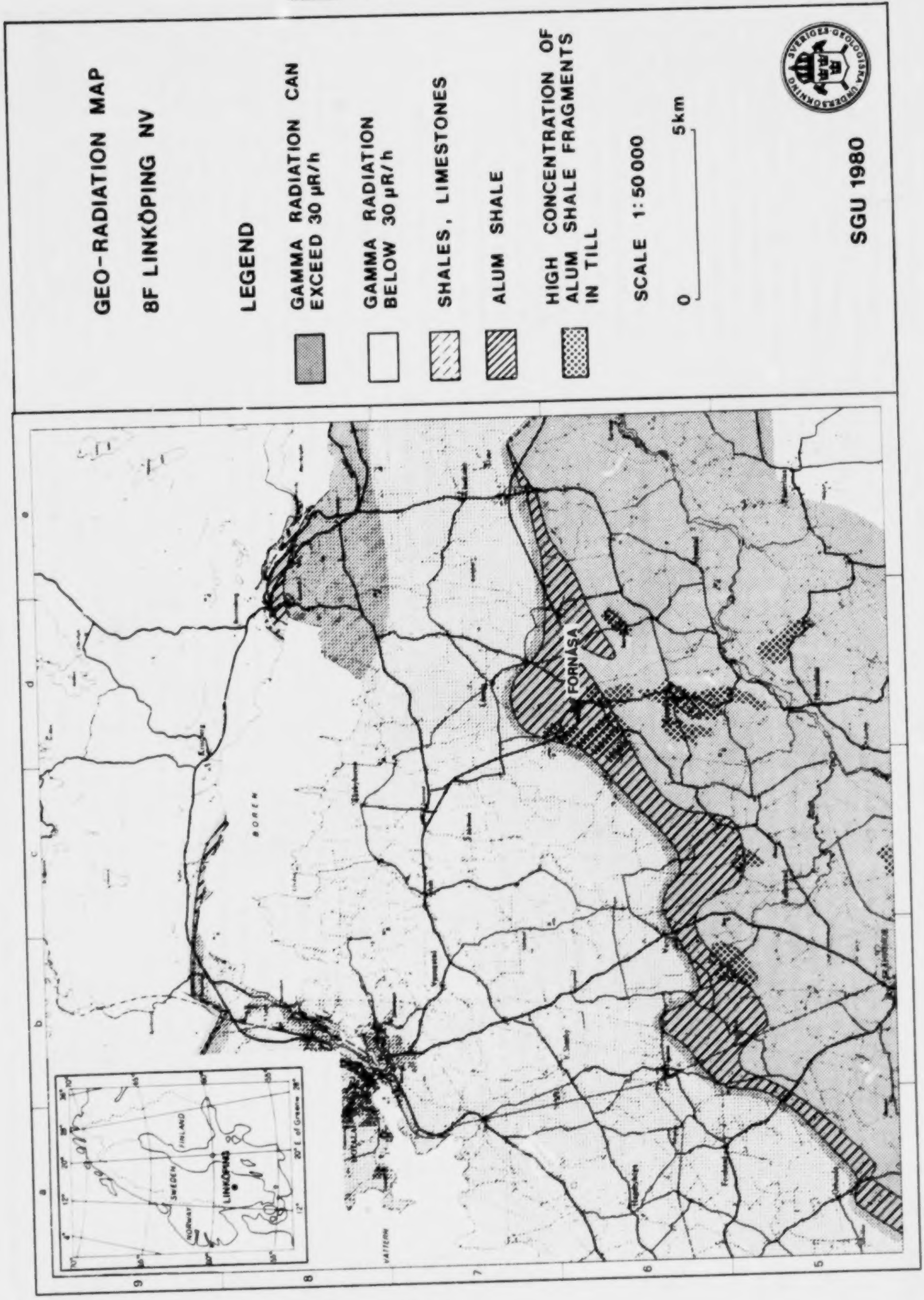
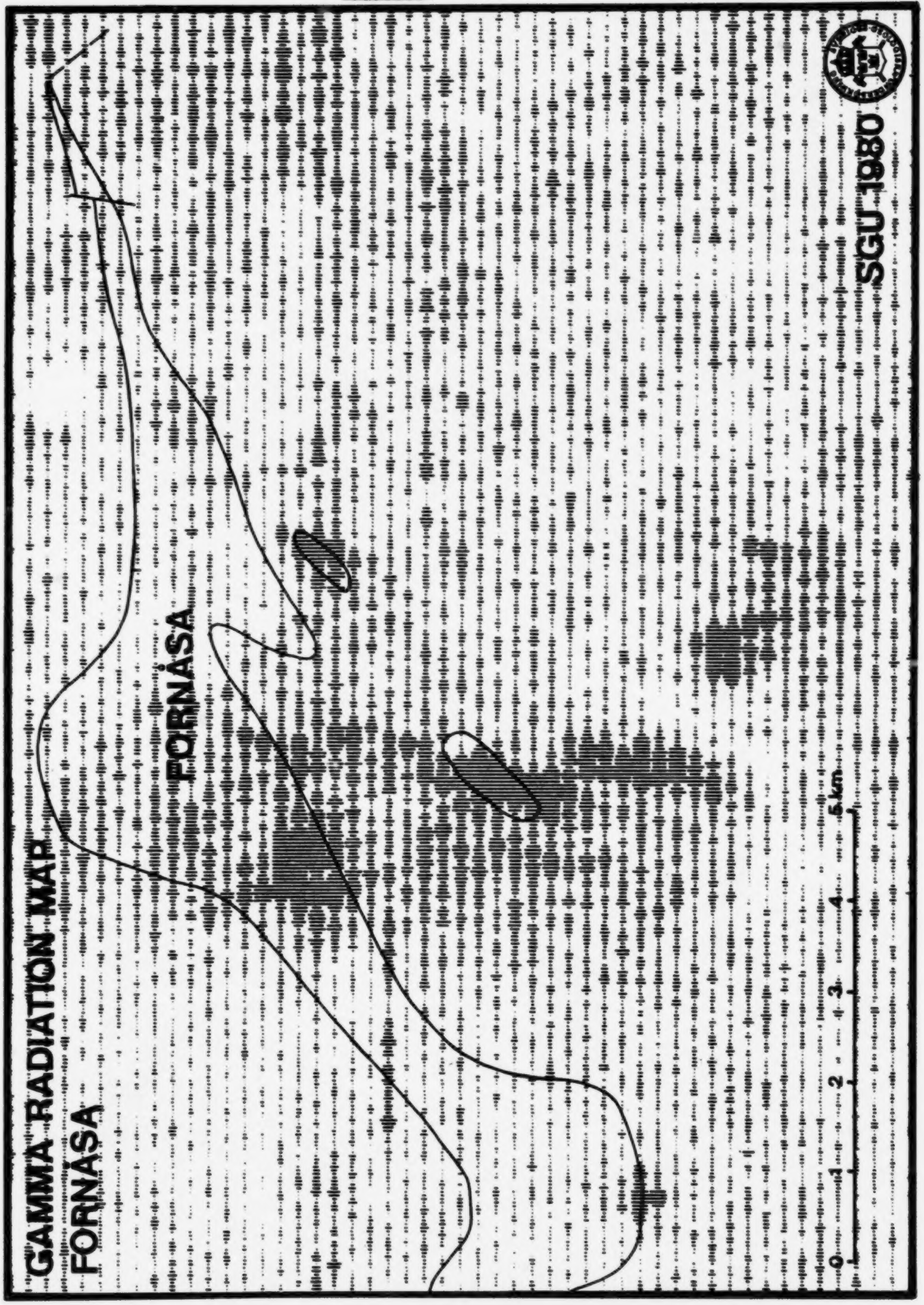

GEO-RADIATION MAP
8F LINKÖPING NV
LEGEND
GAMMA RADIATION CAN EXCEED 30 µR/h
GAMMA RADIATION BELOW 30 µR/h
SHALES, LIMESTONES
ALUM SHALE
HIGH CONCENTRATION OF ALUM SHALE FRAGMENTS IN TILL
SCALE 1:50 000
0
5km
SGU 1980
FORNÅSA
BOREN
VÄTTERN
SWEDEN
FINLAND
NORWAY
LINKÖPING

Figure 6



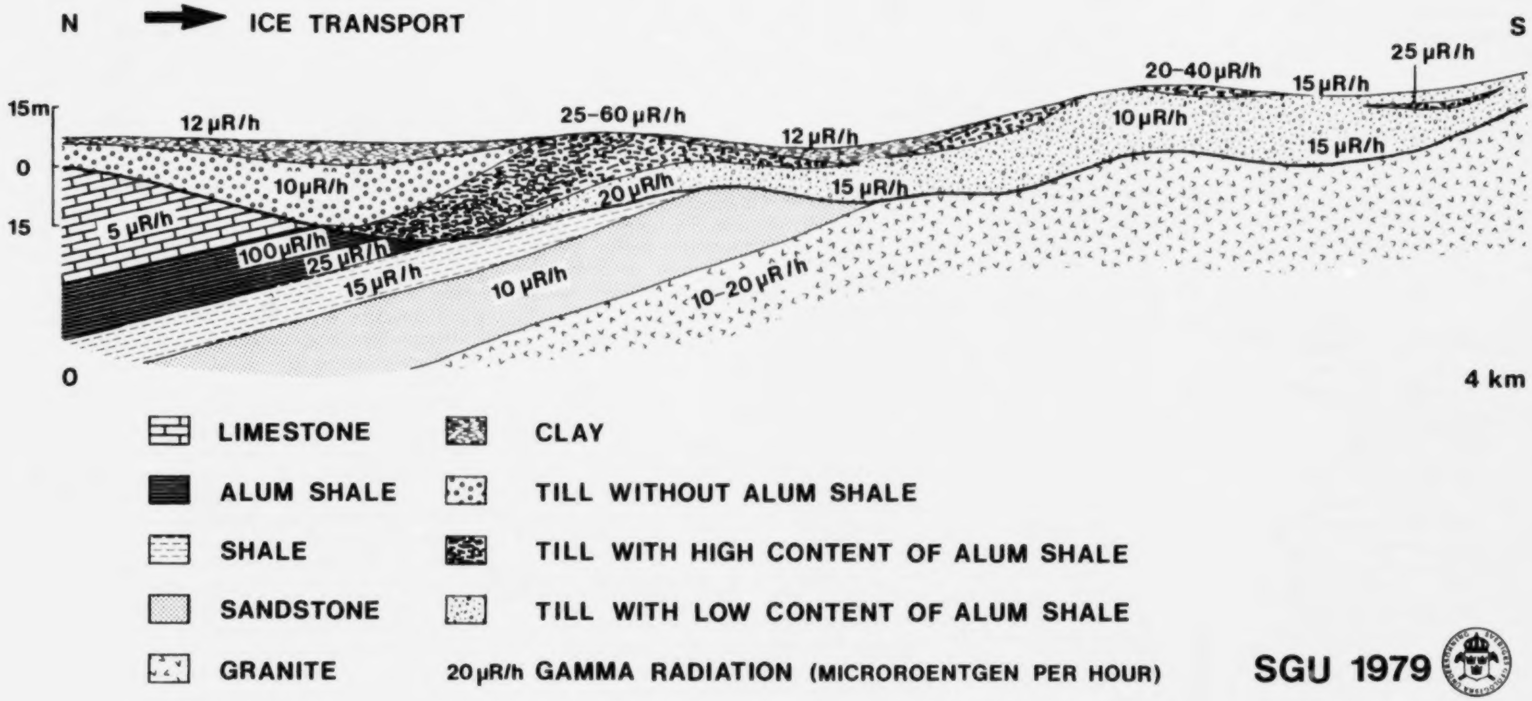
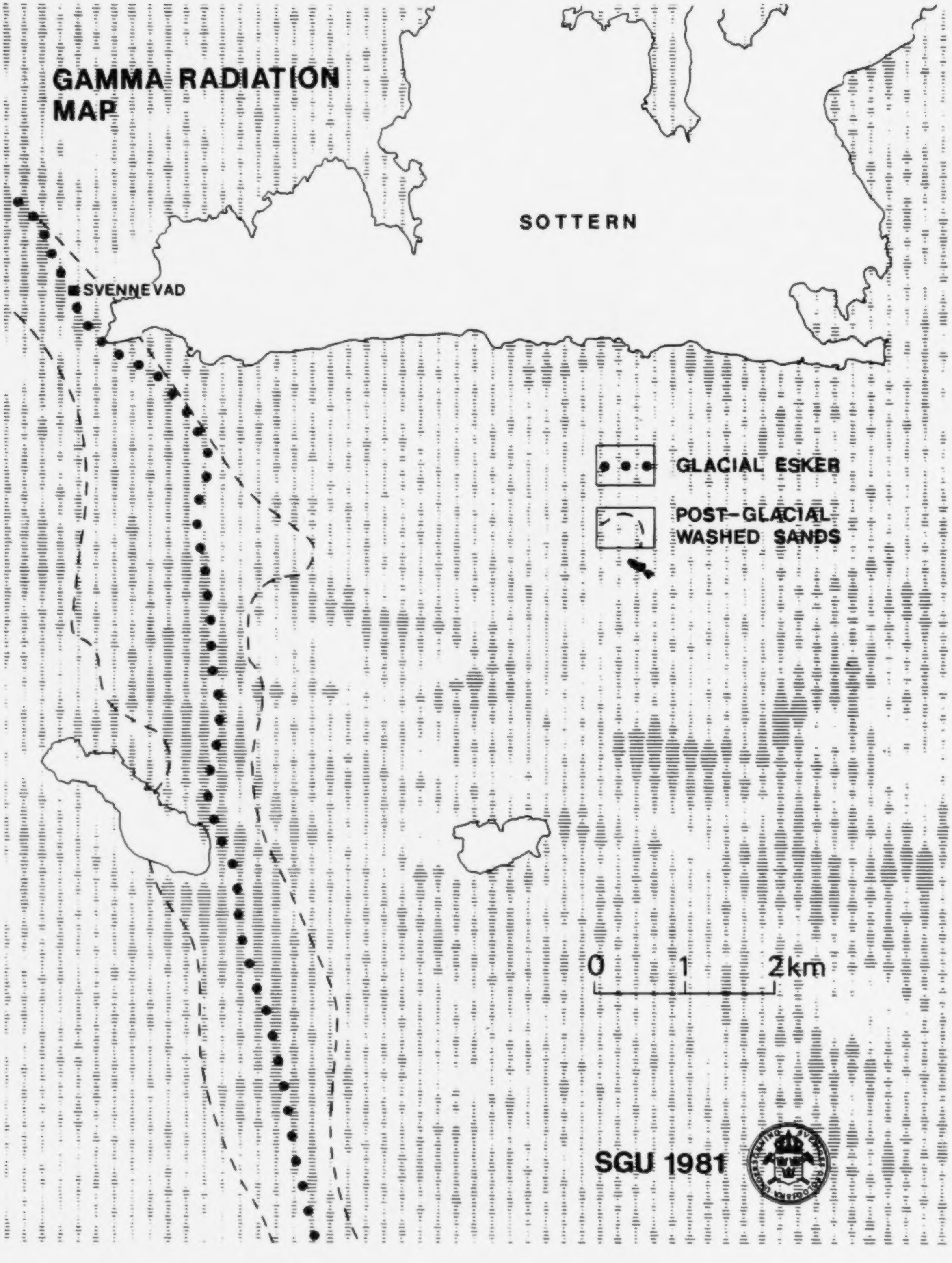

N
ICE TRANSPORT
S
15m
0
15
0
4 km
12 µR/h
25-60 µR/h
12 µR/h
20-40 µR/h
15 µR/h
25 µR/h
10µR/h
10 µR/h
15 µR/h
5 µR/h
100µR/h
25 µR/h
20 µR/h
15 µR/h
15 µR/h
10 µR/h
10-20 µR/h
LIMESTONE
ALUM SHALE
SHALE
SANDSTONE
GRANITE
CLAY
TILL WITHOUT ALUM SHALE
TILL WITH HIGH CONTENT OF ALUM SHALE
TILL WITH LOW CONTENT OF ALUM SHALE
20µR/h GAMMA RADIATION (MICROROENTGEN PER HOUR)
SGU 1979

Figure 7

Figure 8


GAMMA RADIATION MAP
SOTTERN
SVENNEVAD
GLACIAL ESKER
POST-GLACIAL WASHED SANDS
0 1 2km
SGU 1981

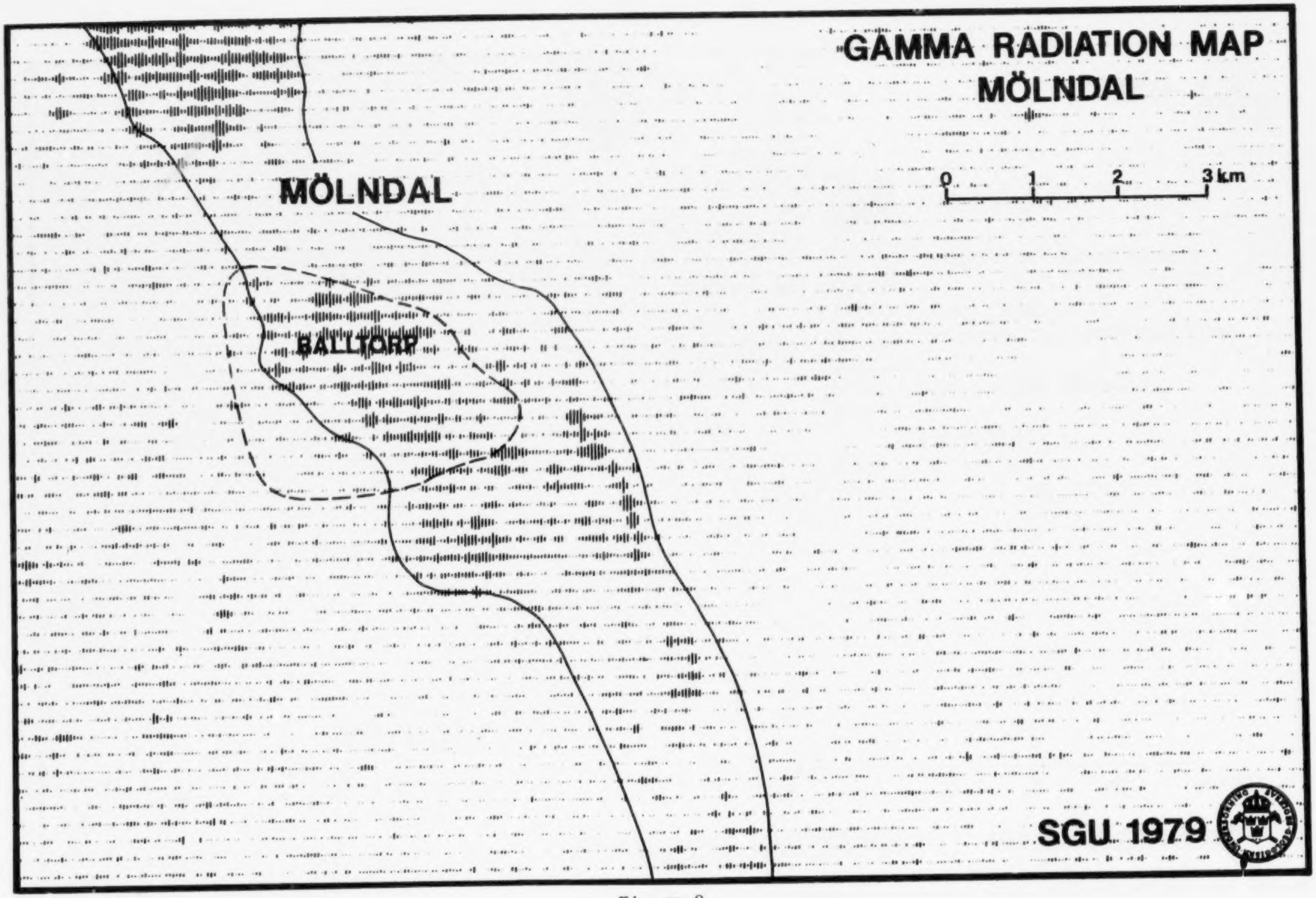

GAMMA RADIATION MAP
MÖLNDAL
0
1
2
3 km
MÖLNDAL
BALLTORP
SGU 1979

Figure 9

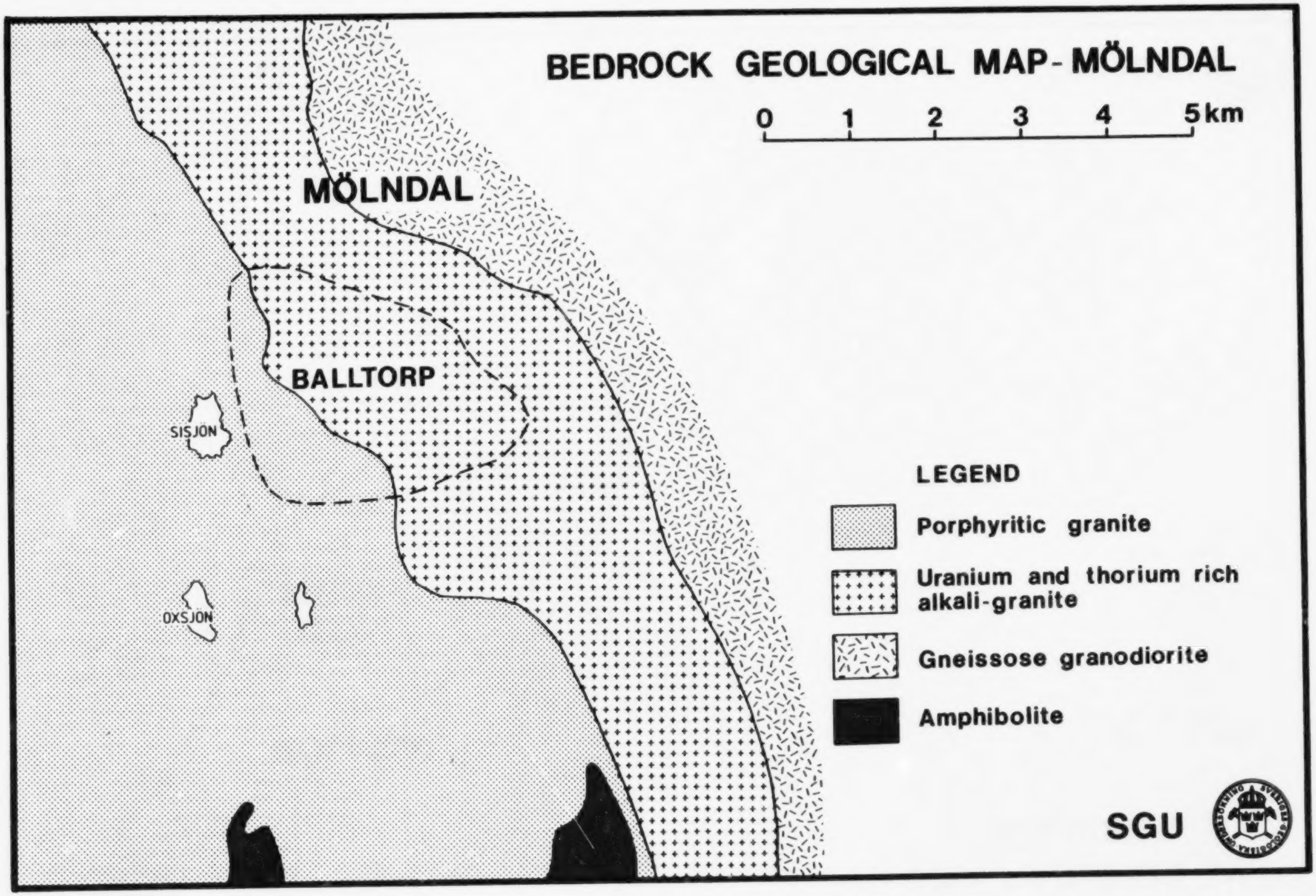

BEDROCK GEOLOGICAL MAP - MÖLNDAL
0
1
2
3
4
5km
MÖLNDAL
BALLTORP
SISJÖN
OXSJÖN
LEGEND
Porphyritic granite
Uranium and thorium rich alkali-granite
Gneissose granodiorite
Amphibolite
SGU

Figure 10

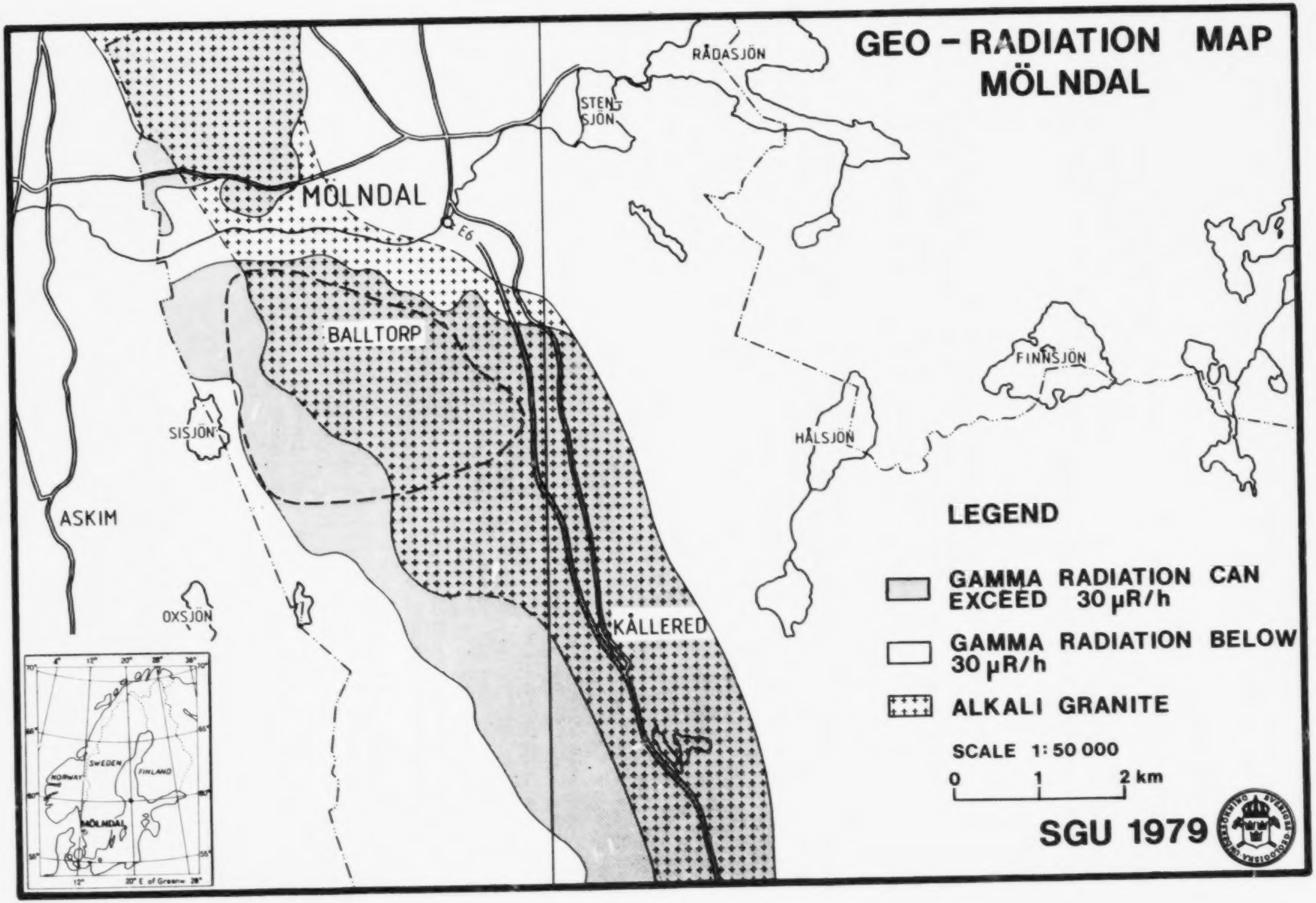

GEO - RADIATION MAP
MÖLNDAL
RÅDASJÖN
STENSJÖN
MÖLNDAL
E6
BALLTORP
FINNSJÖN
HÅLSJÖN
SISJÖN
ASKIM
OXSJÖN
KÅLLERED
LEGEND
GAMMA RADIATION CAN EXCEED 30 μR/h
GAMMA RADIATION BELOW 30 μR/h
ALKALI GRANITE
SCALE 1: 50 000
0 1 2 km
SGU 1979
NORWAY
SWEDEN
FINLAND
MÖLNDAL

Figure 11

A program of radon measurements was carried out in the area during the summer of 1980 (Hesselbom 1981). The results indicate soil gas radon levels ranging from 30,000 to 500,000 Bq m^{-3} (relative values).

RECENT RESEARCH

During the last twelve months, a research program has been initiated to investigate the relationship between radon concentrations in soil gas and radon/radon daughter levels in dwellings, and to test the validity of the GEO-radiation maps. Test areas were selected in areas of both uranium-rich granite (Lysekil) and alum shale (Fjugesta).

Lysekil

In the town of Lysekil, radon daughter concentrations have been measured in 60 selected houses. Seventy percent of these houses have radon daughter levels exceeding the permitted limit for new development (70 Bq m^{-3}, Swedish Building Code, 1981), and 10% have levels exceeding the permitted limit for existing dwellings (400 Bq m^{-3}).

The town of Lysekil lies on the west coast of Sweden in an area of granite known as the Bohus granite complex (Fig. 12). The granite occupies an area of about 20 km by 90 km in Sweden and extends into SE Norway. It has the form of a large flat-lying, sheet-like intrusion consisting of a series of granite types ranging in color from red to grey, and in texture from coarse-grained, sometimes porphyritic to extremely fine-grained (Asklund 1947). Chemically and mineralogically, however, the granite is fairly homogeneous and the whole of the granite belt has enhanced uranium and thorium contents (Wilson and Akerblom 1980). The GEO-radiation maps covering the granite mark the entire complex as having gamma-ray levels exceeding 30 μR/h. In reality, gamma radiation from the ground varies between 10 and 50 μR/h, the lower values in areas where the granite is covered by loose sediments, sands and clays. The higher values refer to the exposed bedrock. Numerous ground spectrometer measurements over the granite give the following uranium-, thorium- and potassium contents: U 9-37 ppm, Th 25-90 ppm and K 3.5-5.0%.

Airborne radiometric surveying has been carried out over the southern part of the granite. Fig. 13, showing the U-component of the gamma radiation map, illustrates clearly the intensity of gamma radiation from the granite compared with that from the adjacent gneisses with normal U and Th contents.

Measurements of radon concentrations in well waters from the Bohus granite give, in general, high levels. In the Lysekil area, 50% of the investigated wells (ca 200) have levels exceeding 1000 Bq/l. The levels recorded compare with those obtained from uranium-rich granites in Maine (Hess et al. 1980). In the gneisses adjacent to the Bohus granite, radon levels in well water are low.

Fjugesta

The indoor measurements of radon daughter concentrations carried out in Fjugesta (45 houses) show that 80% of the houses have levels exceeding the permitted limit for new development and 12% have levels exceeding the

Figure 12

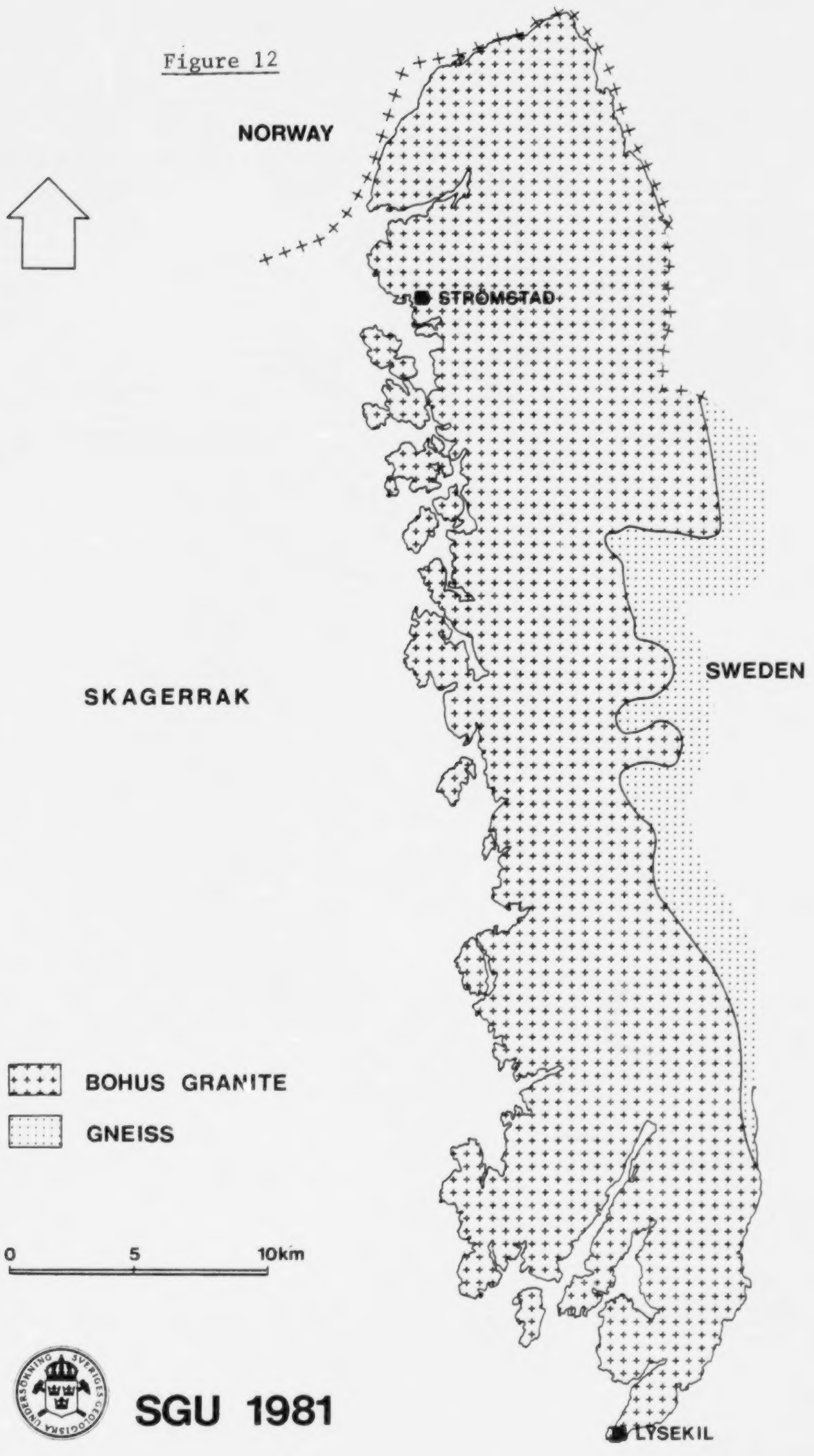
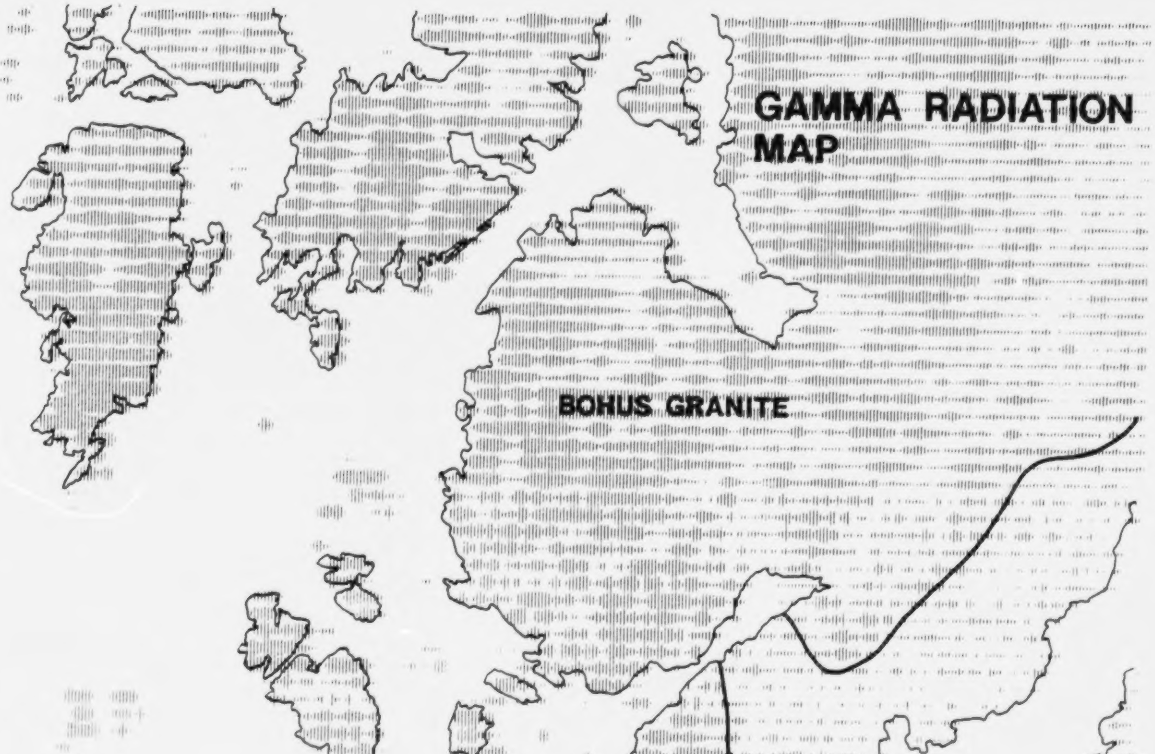

NORWAY
STRÖMSTAD
SKAGERRAK
SWEDEN
BOHUS GRANITE
GNEISS
0
5
10km
SGU 1981
LYSEKIL

Figure 13



permitted limit for existing dwellings. The bedrock of the area, consisting of flat-lying Cambro-Ordovician sediments including alum shale, is largely covered by up to 10 metres of glacial till and post glacial sediments. Fig. 14, showing the U-component of the gamma radiation map and the bedrock outcrop of the alum shale formation, indicates clearly the spread of alum shale fragments in the till. The geological map over the area is shown in Fig. 15.

CONCLUSIONS

The problem of high soil gas radon levels in Sweden is seen now to be acute. Rocks and soils with enhanced uranium contents occur fairly extensively in the country. The normal background gamma radiation in Sweden is 6-10 μR/h, and vast areas have gamma-ray levels of 12-20 μR/h. Investigations so far carried out indicate that the GEO-radiation maps, based on gamma-ray measurements and with an arbitrary limit of 30 μR/h for risk areas, adequately define the areas with particular risk for high soil gas radon contents. However, extensive programs of indoor measurements carried out during the past year by Local Health Authorities and by the National Institute for Radiation Protection, reveal that levels exceeding the permitted limits occur sporadically even in areas where gamma-ray levels for the bedrock and drift cover are considerably lower than 30 μR/h. A number of soil gas radon measurements carried out by SGU in such an area show that radon concentrations in the ground can vary enormously from one measuring point to another. A lower gamma-ray level, for example 20 μR/h, could be adopted for the GEO-radiation maps, but the consequences of this would be that vast areas of the country would be placed under stringent building restrictions which are probably not warranted.

Figure 14

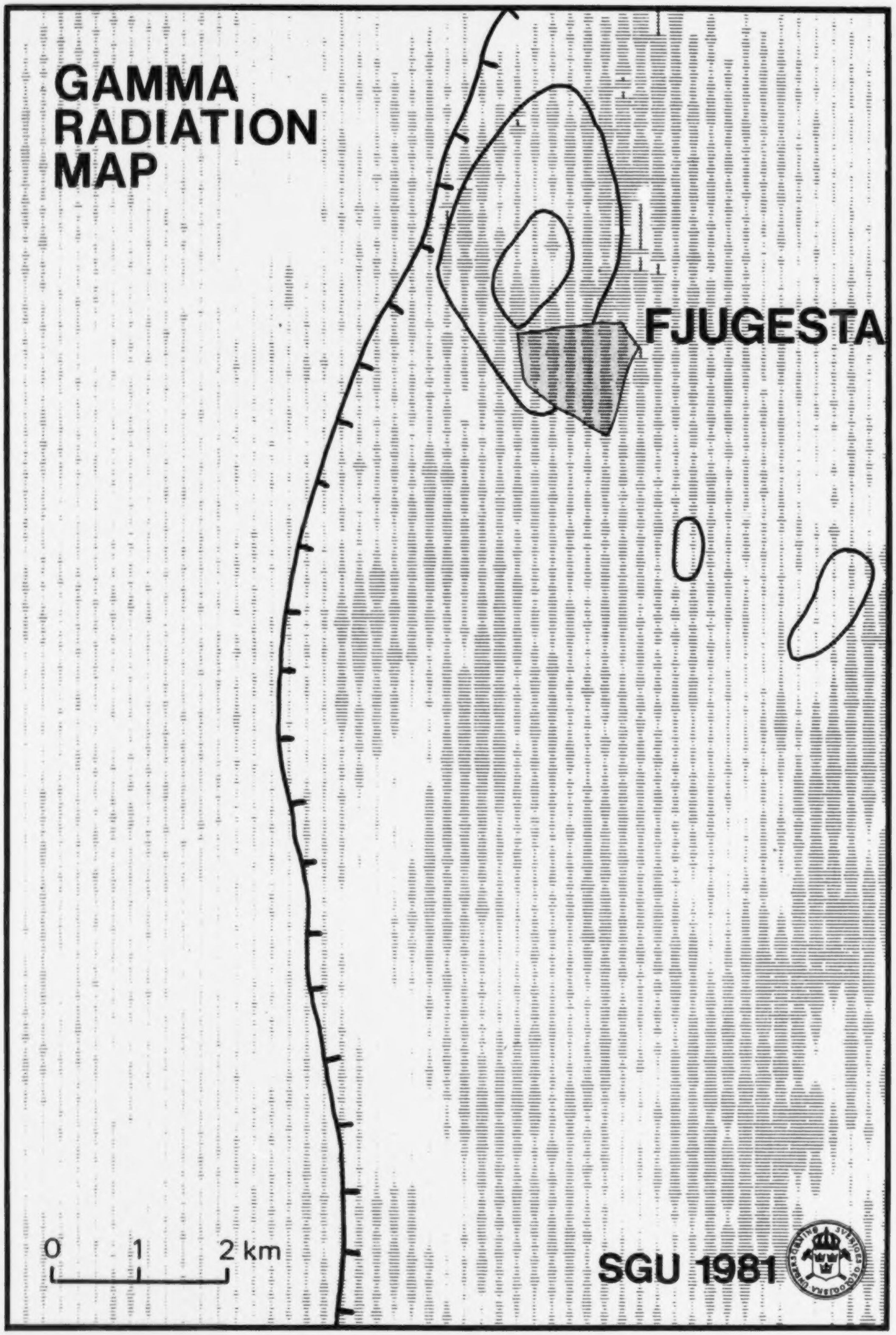
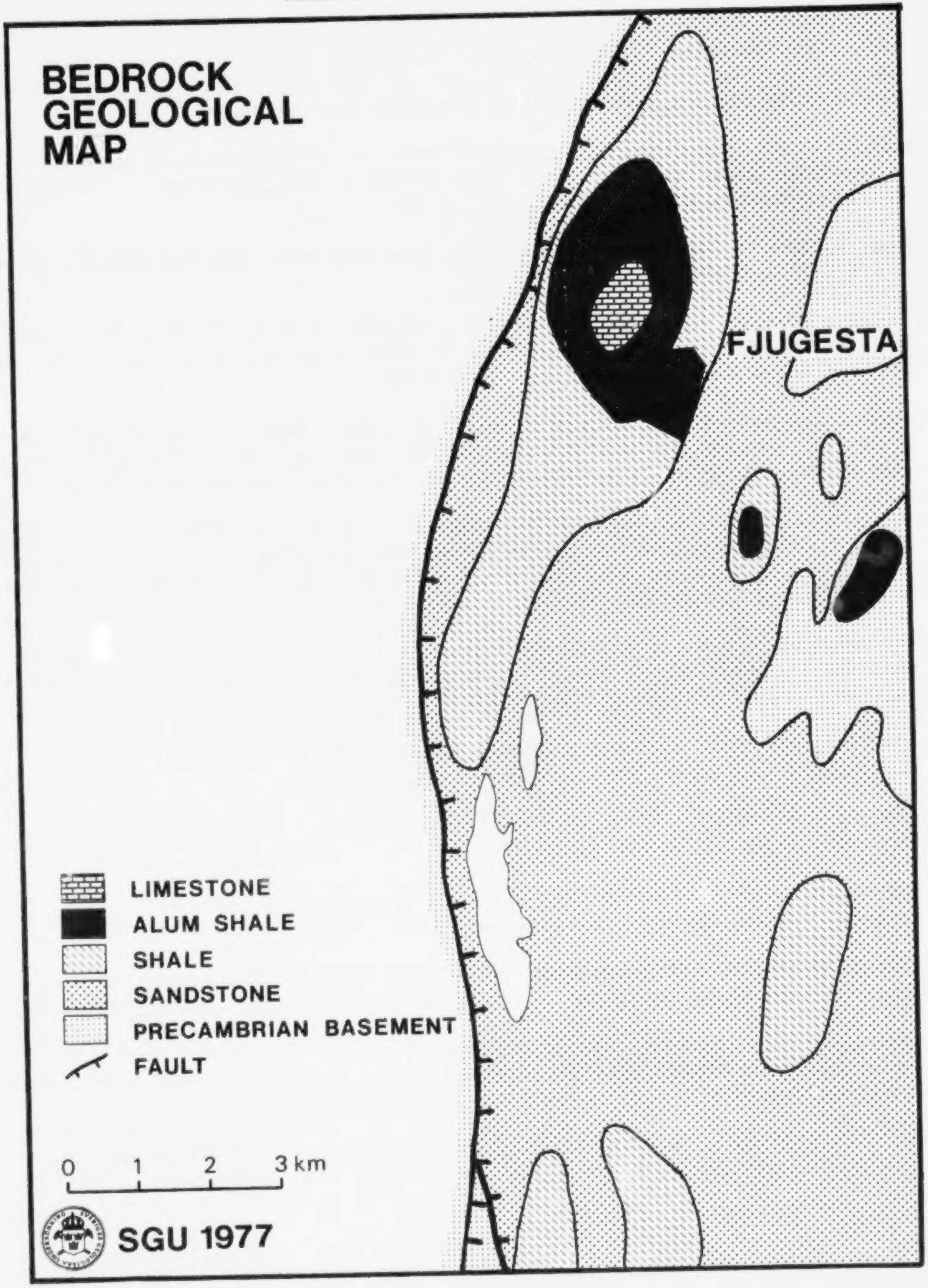


Figure 15


BEDROCK GEOLOGICAL MAP
FJUGESTA
LIMESTONE
ALUM SHALE
SHALE
SANDSTONE
PRECAMBRIAN BASEMENT
FAULT
0
1
2
3 km
SGU 1977

References

Andersson, A., Dahlman, B. and Gee, D.G. in press. Kerogen and uranium resources in the Cambrian alum shales of the Billingen-Falbyggden areas, Sweden. Geologiska Foreningens i Stockholm Forhandlingar.

Armands, G. 1972. Geochemical studies of uranium, molybdenum and vanadium in a Swedish alum shale. Acta Universitatis Stockholmiensis, Stockholm contributions in geology, XXVII (1), 1-48.

Asklund, B. 1947. "Gatsten och kantsten", Sveriges Geologiska undersokning, C 479 (in Swedish), 187 pp.

Hess, C.T., Norton, S.A., Brutsaert, W.F., Casparius, R.E., Coombs, E.G. and Hess, A.L. 1980. Radon-222 in potable water supplies of New England. New England Water Works Association, 2, 113-128.

Hesselbom, A., Israelsson, S. and Tovedal, H. 1981. Radon in the ground. A study of methods and instruments for determining radon concentrations in the ground. Byggforskningsradet, Report R47:1981 (in Swedish), 75 pp.

Linden, A.H. and Akerblom, G.V. 1976. Method of detecting small or indistinct radioactive sources by airborne gamma-ray spectrometry. In: Geology, mining and extractive processing of uranium. Ed. M.J. Jones. Institute of Mining and Metallurgy, London, 113-120.

Radonutredningen, 1979. Preliminary proposals for measures against radiation hazards in buildings. Report from the Radon Commission, Jordbruks-departementet, Ds Jo 1979:9, (in Swedish) 114 pp.

Statens Planverk, 1980. Svensk Byggnorm (Swedish Building Code). Statens Planverks forfattningssamling 1980:1 (in Swedish).

Swedjemark, G.A. 1978a. Radon in dwellings in Sweden. Statens Stralskydds-institut, Report SSI: 1978-013, 1-24.

Swedjemark, G.A. 1979b. Indoor measurements of natural radioactivity in Sweden. Statens stralskyddsinstitut, Report SSI: 1979-026, 1-11.

Swedjemark, G.A. 1980. Radioactivity in houses built of aerated concrete based on alum shale. Statens stralskyddsinstitut, Report SSI: 1980-14 1-6.

Swedjemark, G.A., Hakansson, B. and Hagberg, N. 1979. Radiation levels in houses built on wastes from processing of alum shale. Statens stralskydds-institut, Report SSI: 1979-006, 1-43, (in Swedish).

Wilson, M.R. and Akerblom, G.V. 1980. Uranium-enriched granites in Sweden. Sveriges geologiska undersokning, Rapporter och meddelanden, nr. 19, 1-30.

FIGURE CAPTIONS

Fig. 1. Distribution of the alum shale formation.

Fig. 2. Distribution of known radioactive granites.

Fig. 3a. Uranium-component gamma radiation map over a uranium- and thorium-rich granite near Vasteras, central Sweden. Original scale 1:50,000.

Fig. 3b. Geological map over the same granite. Original scale 1:50,000.

Fig. 4. Geological map for the map sheet Linkoping NW, southeastern Sweden. Original scale 1:50,000.

Fig. 5. GEO-radiation map for the map sheet Linkoping NW, southeastern Sweden. Original scale 1:50,000.

Fig. 6. Uranium-component gamma radiation map over Fornasa, Ostergotland. Original scale 1:50,000.

Fig. 7. Profile illustrating the distribution of alum shale fragments in the Quaternary drift cover.

Fig. 8. Uranium-component gamma radiation map over Svennevad, southeastern Sweden. Original scale 1:50,000.

Fig. 9. Uranium-component gamma radiation map over Molndal, Gothenburg. Original scale 1:50,000.

Fig. 10. Geological map over Molndal, Gothenburg. Original scale 1:50,000.

Fig. 11. GEO-radiation map over Molndal, Gothenburg. Original scale 1:50,000.

Fig. 12. Geological map over the Bohus granite complex, western Sweden.

Fig. 13. Uranium-component gamma radiation map over the Bohus granite around Lysekil. Original scale 1:50,000.

Fig. 14. Uranium-component gamma radiation map over Fjugesta, central Sweden. Original scale 1:50,000.

Fig. 15. Geological map over Fjugetsa. Original scale 1:50,000.

AN APPARATUS FOR CALIBRATING PASSIVE INTEGRATING RADON MONITORS (PRIMs)

Robert A. Washington

Atomic Energy Control Board
Post Office Box 1046
Ottawa, Canada

ABSTRACT

A description is given of a chamber used at the Atomic Energy Control Board laboratory for the calibration of passive radon integrating monitors (PRIMs). It has been proven capable of providing stable and reproducible (to ± 5% or better) concentrations of Rn^{222} in air from about 20 pCi/l (740 Bq/m^3) to more than 3,000 pCi/l (111,000 Bq/m^3). The temperature range employed was 25^o to 30^o, and the relative humidity was in the range from 58% to 62%. The results indicate that a calibration curve of the form $y = mx + b$ (where y = cumulative radon exposure in picocurie-hours/litre, and x is the TLD reading in nanocoulombs) provides better information about PRIM performance than a simple calibration factor $k = Y/x$.

OPEN DISCUSSION I: AUGUST 27, 1981

Open Discussion: Instrumentation and Measurement Methods

Moderator: Peter G. Groer

Panel: Speakers for Sessions I and II

Ron Collé
Andreas Wicke
James E. Gingrich
Howard H. Prichard
Andreas George
Donald J. Keefe
Susan Hinkins
Lyle H. Rathbun
John G. Pacer

PROCEEDINGS

MR. PHILLIPS: I know it's not convenient to go to a microphone every time you have a question, and in a discussion session like this it will be even more difficult. But I think it's important to try to get this portion of the program in the proceedings. We're not only taping these sessions, but we also have a recorder--not a mechanical recorder obviously--with us for that purpose. So it's important, again, that you identify yourself into the microphone. And, for the microphone on my left, again, be sure to speak directly into it.

I made one mistake, one bad mistake, this morning. The paper by Melvin First listed in the proceedings is to be presented by Stephen Rudnick, and it will be presented. I apologize for that confusion.

The moderator for this session is Peter Groer, who you know by now, if you didn't know him beforehand. So, Peter, I'll turn it over to you.

MR. GROER: Well, thanks. For the first few minutes, I'd like to see how things are going. So I'll open the session to the floor, and I think we'll just accept questions.

Please identify yourself clearly. When you use technical terms, try to pronounce them as clearly as possible. It's quite a job to record all of these accurately. I have experience from other meetings; it's very hard to do.

We'll start just with an open question session and we'll see which way the discussion turns.

MR. HOLUB: Question to you. You mentioned the statistics. I remember that Lucas said that if you count the same nucleus twice, once as radium A and once as radium C prime, you have greater error than just Poisson statistics, square root of the count, indicate. Do you have any feeling of how much that is?

MR. GROER: You are correct in pointing to Henry Lucas. He has done very interesting work in this area--together with an applied mathematician at Argonne. The results he has are not applicable to radon daughter counting. I have looked at the problem, and, as I said in my talk, I cannot calculate the variance for the daughter and the granddaughter. For radium A it's very easy. Okay. So, you're invited to try to solve this. I don't know the answer.

MR. HOLUB: In which sense did Lucas make a mistake? Why was he wrong?

MR. GROER: I did not say that he made a mistake. He looked at a different problem. He was not concerned with radon daughter counting. It's the paper--I hope we're talking about the same paper--it's the paper in the Journal of Applied Physics, I think.

MR. HOLUB: Yes.

MR. GROER: He is not concerned with the problem of radon daughter counting.

MR. SCOTT: I was just struck by the fact that literally everyone seems to be concerned with radon daughter counting, with the possible exception of the Germans. What about thoron? I mean, how come we're discriminating against thoron so effectively? Is thoron a problem or do we just ignore it?

MR. GROER: I think that's a very good question. If there's anybody here on the panel who would like to answer this, be my guest. All I can say is that I made many measurements in the U.S. in uranium mines and in other locales--then spectroscopically--and I've never seen thoron daughters. But I know that it's a problem in Canada. And I am sure there are people here who know what the thoron daughter concentrations are in Canada. David, to this point?

MR. ROSENBAUM: I think the basic policy reason and the reason in the ORP why we haven't done anything on thoron is that the health effects data are all in terms of radon daughters. And, therefore, if you want to know the effect on somebody you look at the radon daughter concentrations, the working levels, and from that you induce the danger, the risk.

Now, the implicit assumption here is--and that's because the original measurements in the mines threw away the thoron--that either thoron is so small that it isn't a problem, and therefore doesn't make any difference, or the ratio of thoron daughters to radon daughters is the same as it was in the mines. And, therefore, when you throw it away the health effect still comes out right. Undoubtedly, there are situations in which both those assumptions are completely wrong.

MR. SCOTT: If I could just carry on with this. One place where it does strike me as being particularly important (I may point out, I am from Canada and there isn't all that much thoron around, but it is present.) is in the kind of instrument that does estimates of radon daughters by, say, alpha decay or possibly even alpha spectroscopy plus beta counting. The resulting estimates will be severely distorted with even the presence of a small amount of thoron, particularly in the beta plus alpha ones.

MR. GROER: No doubt about it, if you have thoron daughter concentrations that are high enough, they will, since they fall into the two different alpha channels, the upper alpha channel, there is no doubt about it that this will distort measurements. So the instrument you heard about today cannot be used in its present form in atmospheres where there are thoron daughters. I agree with you.

MR. PERDUE: If I might make a comment on the thoron. Of course, there is another radon we haven't talked about, which is 219. But anyway, I think we'll all agree that in the world in general, thorium, in background levels, is larger than the amount of radium there is in the soil, quantitatively, or by weight. So if one takes measurements approaching background levels, there has to be thoron in it. And you can measure this.

MR. GROER: Well, there's one big difference in the two decay chains. That's the half life of the noble gas, right? And that makes a difference. So it depends very much on your situation. I don't dispute the existence of it, but radon-220 does have a short half-life.

MR. PERDUE: That is easily explained. Radon-222 may decay in the next state, but radon-220 decays right where you are.

MR. ROESSLER: Just one small comment on the thoron question. In the heavy mineral industry, in particular I'm looking at zircon mill and products, using two total alpha counts at different times--I guess, a rock--we did find both radon daughters and thoron daughters. In fact, if you calculate in terms of working levels, the thoron daughter working level of that particular case is slightly higher than the radon daughter working level. But the summation of the two is still several orders of magnitude below occupational standards, so we never really followed through on that in any great detail. But there are places where we're going to find thoron daughters, yes.

MR. GROER: Okay. Any more comments on the thoron and thoron daughter question?

MR. ROSENBAUM: The National Radiation Protection Board estimates that there is about ten percent as much thoron as radon in a typical house. I don't know if that is peculiar to Britain or the building materials that are used or what it's from, but that's the kind of numbers that they use. And they might be enough. I'm no kind of instrumentalist, but that might be enough to interfere with some of the measuring techniques.

MR. COHEN: One last comment on thoron. My lab is in the United States, and there's plenty of thoron in it. In fact, it puts a fair correction on the radium A peak, enough to really make a difference in our lab at least.

MR. GROER: What causes the correction to your radium A peak? Thoron?

MR. COHEN: There's a thoron alpha that is the same energy as the radium A.

MR. GROER: Yes, but why do you think that you have thoron in your filter, if you use a filter method?

MR. COHEN: Because there's also a thoron peak at 8 point--8 MEV. But there's also a thoron daughter at 6 MEV.

MR. GROER: I agree, but not that it's thoron. I mean, it was technical. It's just a little difference. You said thoron and I said no, but that it's a thoron daughter I agree with you. In the houses where we have used this instrument and in the uranium mines with the channel analyzer underground, we have never seen a thoron daughter peak at higher energies than, well, above 7.68 MEV. What is the alpha energy? I think it is 8.7 or something like that. Our highest flow rate, as I mentioned to you, was 40 liters per minute. But we are sampling only for two minutes.

So, if there are no more questions on the thoron daughter, I'd like to steer the discussion a little bit. I want to bring up the question of availability of commercial instruments--how satisfied people are. I was asked, you know, to bring this up. How happy are investigators with commercial instrumentation? What are the problems? What instruments should be produced commercially? Is there a need for a certain type of instrument? Is there any interest in this topic?

MR. HOLUB: I'd like to mention the problem of acquiring a constant radon source one can rely on for these calibrations. And we have some commercially available dry sources too--EPA brought some. We have had bad experiences; some people have had better experiences. Now the onus is really on NBS to produce something really good. The idea is to provide a known, reproducible radon concentration at any time. So, I wonder how interested others are. We certainly are. I think it would be useful to have them.

MR. GROER: If I understand you correctly, you're calling for a radon meter that would measure--

MR. HOLUB: Radon source. Always produce a given amount, hopefully a solid source, so that you don't spill it. The more I question, the more I think NBS is going to do it.

MR. COLLÉ: Well, yes--

UNIDENTIFIED SPEAKER: Stop right there. Good concise answer.

MR. COLLÉ: The onus may indeed be on us. What he was referring to, of course, is some type of solid source to replace our solution standards. Although there are laboratories that can use solution standards--use them very well, perhaps better than we can--there is a real need out there for a solid source, something like the commercially available source, which would be very easy to use. We have a very limited effort underway looking for a replacement in a solid source. And, if the onus is on us, the last comment I'd like to make is, if you see me, I'll tell you who to make the check out to.

MR. KAHN: What about a liquid source with two valves on it so you can bubble? What we need is some source that can be used over and over again. Is there any problem with that?

MR. COLLÉ: Well, we had a small meeting, with a few people, some EPA people, and for such a type of source, we discussed several types of configurations. One of them was a solid source like ion exchange resin impregnated with radium, in which case somehow magically either the radon would get out of this material into a solution and then you would take it out of the solution, or perhaps take an aliquot of that solution for making radon in water analyses.

We have considered about three or four different designs, none of which have been excluded yet, and your type of proposal is being considered. I must say that this effort is relatively low key, but they are being looked at. There are several possibilities and that which you've mentioned hasn't been excluded.

MR. PHILLIPS: Since I suggested, Peter, the question about the commercial instruments, I feel compelled now to try to continue comment on that. I have not used the newer commercial radon daughter instruments, but I have looked at a few of them and, just from initial observations, it appears to me that there may be some problems in the intake portion in terms of plate out, prior to deposition of the daughters on the filter. I know in particular you were concerned about this for the instrument that you and Don worked on. I wonder if there is any other comment on that, particularly from you.

MR. GROER: As you probably remember from the figures you saw on the slides this morning, we tried to keep the air inlet completely unobstructed because we were worried about the so-called umbrella effect, that we are losing the aerosol particles on possibly the detector that shields the air. Now, I do know that there are commercial units around that measure radon daughter concentration, or I should say total alpha counts, rather than individual radon daughter concentrations. Very little care has been taken to avoid plate out problems. I have seen an instrument where the air inlet is quite a small opening, and then a few inches away from that is the filter. There is just no doubt in my mind that you will lose some of the radon daughters on the way to the filter.

So, I think this is an important problem, and I think manufacturers should be made aware of that. Unfortunately, as long as there are customers who buy these units, the manufacturers will not be forced to change their assembly lines.

MR. KEEFE: I'd like to make one comment with regard to the problem of plating out on the detector. From an engineering standpoint, one of the problems that you have is that it's easy to design an unobstructed air intake port. The basic problem is to get the filter membrane from point A to point B, and this is the area that costs a lot of money. This is also where your mechanical problems lie. I'd say 30 percent of the cost of our instrument lies in that particular problem. Industry is going to have the same problem, except they're looking for a product that is lower in price, that has a good market and saleability, and they're trying to cut in that area, I feel.

But, if the scientific community and the people who are interested can bring out the point that plate out is a serious problem and they should consider these things even though the costs of their instrumentation may go up, I think they may make some changes in their instrumentation and they could solve that problem.

MR. NYBERG: We have looked at--in our lab--quite a few instruments, not as many as Andy George, but then I don't think anyone has. I think as an electronics engineer I can say, speaking, I think, for some of us, that the instruments are getting better. Just in the last few years we have seen an awful lot of newer, better, smaller, lighter, more versatile instruments coming out. One of the problems that we have that maybe could be addressed by this group is how do we evaluate them? Now we send them to Andy or some such thing. This business about plate out on the detectors is a very common problem. There's a much too tortuous path for these daughter instruments. But there is not even a semi-standard way of evaluating this at the present time. I'd like to see if someone has some suggestions for looking at these things in a little more organized fashion than we're doing right now.

MR. GROER: Thank you. I think that's a very good point. It also leads to the question of intercalibration and things like that. I'm sure that some of our panelists here will have some ideas on that.

MR. COLLÉ: No, I don't have ideas on it. I just want to reinforce the point and perhaps give you something else to think about. What we have in this room is a collection of probably the premier laboratories in this area.

What you have to keep in mind as well is that most of the radiation protection being done in this country is being done by state laboratories, and the fact of the matter is that their instrumentation needs are considerably different than research laboratories' needs. They need instruments that are very reliable and relatively inexpensive, particularly if they have many sites to cover. They cannot spend 10 or 20 thousand dollars for an instrument.

Another matter related to calibration and the reliability of instruments is that there really should be some type of evaluation of instruments to guide some of these other laboratories. These laboratories are not equipped to do the type of evaluations that your laboratory may be able to do, and it would be a great service to them to have this evaluation. Also, there is a great need, once an instrument and/or method is evaluated, to assure that it's compatible, on a continuing basis, with other things.

These are just additional thoughts that I think we should keep in mind. We shouldn't overlook that there is a bigger user community out there than just the people in this room. Now, I will leave the floor open for the ways to do that.

MR. KEEFE: Speaking of calibration, again, and the problems, and state agencies. I have received calls from several state agencies with regard to calibration of the EWLM, which belongs to the EPA, which they have loaned out to some state agencies. They are presently involved in one specific case in the state of Montana in lawsuits versus various cities and mining companies and so forth. Immediately they get involved in having to prove the calibration. The measurements that are made with this instrument must be documented and proved. Again, you're back into the calibration problem.

One of the problems I'd like to mention about calibration, and it was mentioned before, is that people have been using these radon chambers, I hope basically for calibration of radon instruments, not necessarily radon daughter instruments. One of the problems that you have in these large bags is that the volume of gas that's contained inside the bag is not sufficient for our calibration. What we have to do in all our calibrations is go to the uranium mines. We have to go to the Bureau of Mines research mines and use that large volume of air and try to keep the air as static as possible to make our calibration.

Certainly the scientific community could use some sort of a calibration laboratory where it wouldn't have to go that far. Try that in February sometime--in the snow. It's an interesting experience.

MR. GROER: February in Colorado is okay if you ski down. With this, maybe we can leave this topic, or is there further comment? I don't think we really have solved anything. What I personally don't see is how we could get a national quality standard for instrumentation like that. Okay, why not? Let's hear your ideas.

UNIDENTIFIED SPEAKER: I don't have anything more to say. I just wondered why not.

MR. GROER: I just wanted to hear some suggestions. I mean, who would you suggest to be the supervisory agency? The National Bureau of Standards? Some other agency?

MR. PERDUE: I'd just like to say that we've been here most of the week making intercomparison studies of existing instrumentation, and I feel that unless a lot more of these studies are made we're never going to really solve what's going on. There's a number of problems that haven't even been approached. What happens with humidity? Does this change the radon daughter attachment? What happens to the other isotopes? We know sometimes they are indeed present. So, unless we can address first the things we know, we can get Bureau of Standards radium that can be calibrated and, of course, can go back to what they've got. Can we really know what's going on unless we keep this thing up? We are, each one of us, working in a little pocket and we're getting data and we don't know whether it matches somebody else's data or not.

MR. COLLÉ: I think the important point is that it's very admirable that these things keep up. But the problem is this. You have an informal group of six or seven laboratories--and perhaps these six or seven laboratories can answer some of these questions for the other laboratories--but what do the other 30 or 40 laboratories who are also making these measurements do?

MR. RATHBUN: It occurs to me that a lot of the other work like this that's being done has been farmed out to independent companies. And it seems like at this point there is no national agency that wants to be the arbiter for such a project. Why not have an independent group bid on a contract to do this type of work and let each person that wants something calibrated pay a certain fee to bring it in?

MS. FISENNE: I'll help you out on that one.

MR. GROER: We should have comments on this one.

MS. FISENNE: All I can say to that is that's why we have the National Bureau of Standards. So, if somebody is willing to come up with contract money, they will obviously address this problem. I don't think it has to be done as a private industry project. Obviously, the National Bureau of Standards is exactly that, at least in this country. They provide us with our radium standards right now. So if everybody is that much interested, then we should be able to get the money together to allow them to remain again on national standards in this area as well.

MR. COLLÉ: Thank you. That's indeed the point. We have had this type of initiative literally proposed for perhaps a little longer, but definitely for three years. It has ranged from a couple of hundred thousand dollars to multimillion dollar efforts proposed from every possible point of view that you could perhaps think of. You know, we've tried to beat the bushes in all sorts of ways. We thought we hit all the right things and everything else. We have not been successful yet.

We recognize there is a need out there for basic national standards for these measurements. We would like to make an effort to try to provide this. There are probably other laboratories which right at the moment could do a better job of it, but by congressional fiat we have the national responsibility to maintain these things. We have both the long-term interests and commitment to maintaining these things. And I can't agree more with Sue that it should be done on a national basis. That's why we were established as this type of laboratory. And it just takes more of a push than we've been able to provide ourselves.

MR. GROER: Great. What could be the first step?

MR. HOLUB: To make a list of problems and agree on that.

MR. GROER: Can you be a little more specific?

MR. HOLUB: For instance, as I said before, the plate out, the charge, the trace gases, and the humidity--these I consider a beginning--and then the techniques of spectroscopy versus total alpha and error propagation, perhaps in more detail. What happens if the concentration changed during the sampling and it's simply error propagation?

MR. GROER: I agree with you. It is an important research problem. But I think we just addressed slightly different questions. The question was about intercalibration, standardization, possibly of instruments. My concern is how do we take the first step as a group if the National Bureau of Standards is the agency that would be entrusted with such a venture?

MR. COLLÉ: I'm not sure. I'm not sure of the mechanism. I just know that it requires money and personnel, both of which we are drastically short of. How the money comes is not our most important concern. We would certainly not turn down a multimillion dollar contract or interagency agreement. We would prefer direct funding to our own appropriations since that's more stable and one can build continuity in programs. But we have some plans. We have had these plans for a number of years, and it's just a question of putting them into our program and funding them. The mechanism to do that is to have Dr. Rosenbaum write us out a check.

MR. GROER: Well, that was certainly a clear answer. I think Bob Holub addressed some interesting questions this morning, and he just reiterated some of them. I'd like maybe to take up the first one. What do we know about radon daughters immediately after? By immediately after I mean, maybe a picosecond or maybe a nanosecond after they were formed in vacuum. What do we know about that? What charges are there on these, and then what happens from there on? Would anybody like to tackle this question?

Well, there are papers in the literature that tell you exactly what the charge states are for a radium A or for a thorium A. But this is all measured in vacuum--not the typical condition. And then, of course, the neutralization of these daughter products after they are formed is an area that's very poorly understood.

I think there's a research project going on at the University of Illinois. Does anybody knows about this project? Is there somebody here that works in this area? Bob?

MR. HOLUB: That's the project that was funded by the Bureau of Mines, and it has been stopped, not for lack of funds. But the basic facts are that usually people assume that 90 percent of all daughters in the air are positively charged. Now, in 1914, as I mentioned before, Welish found that the few have ether, lots of ether; he obviously didn't have a hundred percent, but lots of ether in the atmosphere, (and) none of the daughters was charged, none. In the arrangement similar to what Andreas Wicke has shown there simply was no correction due to charge.

MR. GROER: Excuse me. Can I interrupt you? This reference will have to go into the record. Could you possibly share it with the group? I think it's an interesting reference, which I personally didn't know.

MR. HOLUB: Yes. Most people reference it, but no one reads it. Unfortunately, I don't remember the full reference, but I can give it to you tomorrow.

In the experiment done in Illinois, published in January in Science, they did exactly what Jess Thomas did, except instead of air passing through they had nitrogen with various trace gases. And when they had 10-ppm and NO_2, the diffusion coefficient was such that it must have been neutral. It was, by the way, about twice as fast as when it was charged like impure nitrogen. It's a very simple and easily understood experiment.

The Canadians did a similar experiment (published in Physics in March), and they had pure gases. When their ionization potential was lower than argon's their results basically confirmed what Hopky found at the University of Illinois. Instead of trace gases, they simply used other gases, like acetylene and formaldehydes and CO_2 or something--I forgot, there are quite a few gases. They all have lower ionization potential than argon; however, they still have higher ionization potentials than polonium.

In Bombay there is an Indian group, Cotropi. They looked at ether. Apparently, besides the ionization potential, the ion molecule reaction also plays a role. All these ion molecule reactions are taking place within a period of microseconds. And, by the way, some people use the term "born neutral," even though, of course, it's not very likely that the daughter is neutral and is slowed down.

So, that is a very difficult problem. And it depends on trace gases. Now, the number of hits per molecule per second is ten to the ninth, so it's basically, if you are ten to the minus ninth of something in the air, still it's going to be a hit once a second. So you can imagine the terrible problem this might be. Even though, as you all say, for practical purposes maybe it's not necessary, but it should be looked into. That's the review of what happened in this in the last half a year or year.

MR. GROER: Okay. So, if I interpret the situation correctly, there is no systematic research effort in this area--neutralization of, let's say, newly formed radon daughter products in the atmosphere, whatever this means.

MR. HOLUB: That's it exactly. We are trying to do something, but it is very difficult.

MR. GROER: Is David Rosenbaum here? No? Well, this is certainly, I think, an interesting question. Humidity was brought up. Now, there are many, many different questions related to humidity. Calibration, filters, TLD, track etch, plate out, I don't really know where to start. If anybody here, the panelists, the audience, has a particular humidity problem he would like to bring up, please do so.

MR. COHEN: This question is related to what you just said. If I understood Jim Gingrich this morning, he said that what a track etch detector measures is what is plated out on it. Now, if that's true, then it must be sensitive to what the plate out conditions are. And, if plate out conditions are sensitive to humidity and all these other things, how can they just be a measure of radon, the radon daughters?

MR. GROER: Jim?

MR. GINGRICH: The current type of detector we have now will measure whatever is plated out on the surface. But for precise radon measurement, Bernie, we use that type F cup. All right, what it does is filter out any radon daughters that are outside the cup. So the only thing that gets through the filter is radon itself. Radon itself will then go into the cup, plate out on the walls and sides of the cup, and plate out on the detector's surface itself. You'll get some signal from the radon that's still in the cup and some signal from the plated out materials on the side of the cup. Does that answer your question?

MR. COHEN: It's a clear answer, except it doesn't agree with things I've heard. I've heard that, for example, if you put a solid state detector where the detector is, you get a clean spectrum. It's as though everything that is being detected is the stuff that is actually plated out on the surface. And if that's true, then even in the cup you're measuring stuff which plates out on the surface of the detector. Am I wrong somewhere?

MR. GINGRICH: Oh, yes, you are measuring stuff that plates out on the surface of the detector in the cup, but you've got to remember that the only thing that will get through the filter is radon. So the signal that the detector sees is directly proportional to the total amount of radon that has come through the filter.

MR. COHEN: The radon comes through the filter and is in the cup. All right?

MR. GINGRICH: Okay.

MR. COHEN: Then it decays into daughters and, as I understand it, essentially what happens then is only the radon daughters which plate out on the detector are detected.

MR. GINGRICH: No, that's not correct. It is seeing those radon daughters as well as the alpha particles from other radon daughters that would have been plated out on the walls of the cup--and also a signal that is directly from the radon itself because the detector does see a certain volume of radon gas.

MR. COHEN: But what's the support for this? I mean, is it true that if you put a solid state detector there you get a clean spectrum with only peaks? That's what I've heard.

MR. GINGRICH: I'm not an expert on solid state detectors.

MR. COHEN: Well, how about with your tracks? I mean, are all your tracks the same? Do they have only two different lengths?

MR. GINGRICH: No. You'll see a full spectrum of tracks. And the reason you will is because if you look at the volume, first, the important thing is where do the alpha particles start within the volume? All right. If it starts some distance out, then by the time it gets to the detector the track that you see in the detector will be fairly small compared to a track that would have started from an alpha particle fairly near.

MR. COHEN: That's what I was asking. You actually have data looking at the length of tracks, and you found that the tracks do have all different lengths?

MR. GINGRICH: Oh, yes. It's a full spectrum.

MR. PHILLIPS: What you said is true; at least I have done that. If you put a solid state detector in those cups, then what you see is a clean A and C prime spectrum. But I think you have to consider the conditions and the solid state detector construction; it has a bias on it, too. So that may not be directly relatable to the track etch situation.

MR. HOLUB: We have done this, too, and the detector on the surface is neutral. The bias probably would not play a role if the daughters were not charged. But there is no bias on the surface of the detector.

MR. PHILLIPS: That's true, if it's operated at a positive potential with a negative potential or ground on the outer surface of the detector.

MR. KAHN: Well, what does that say about the radon? You also see the radon alphas.

MR. PHILLIPS: No, I didn't. In the counts that I did, and these were integrated counts over fairly long periods of time--about an hour at concentrations, let's say, of 50 picocuries per liter, equilibrium around 25 percent--I saw essentially no radon in the spectrum.

MR. KAHN: Can you explain that?

MR. PHILLIPS: My explanation--

MR. GINGRICH: Well, I would agree with you. There's actually a paper that's published on this, and I could give you a reference to it. It's by Dr. Bob Fleisher and tells the contribution of radon and radon daughters, the alpha particles, where they plate out, and so forth. But, as I remember, something like 30 percent of our signal in the cups, in the normal geometry, is due to radon, and the other 65 percent is due to plate out on the side and on the detector. I can check those numbers for you, but I can certainly give you a reference to it.

MR. KAHN: But that doesn't seem consistent with his observation.

MR. GINGRICH: I don't know how to explain that inconsistency, I'm sorry.

MR. PHILLIPS: There's another interesting situation that I haven't been able to explain. (I might add that all of this is preliminary work; I have not followed up on it because I got busy with other things.) If you put it in a filtered cup, you see a clean spectrum, and the heights of the A and C prime peaks are about the same. If you remove the filter at the same concentration, the C prime peak is roughly twice the height, or as many counts, as the A. Somebody tell me why.

MR. GROER: Bob?

MR. HOLUB: In answer to the first question, it depends simply on how big the cup is. If you see the radium A and C prime from the walls, then, of course, you get contributions; it's not a clean spectrum. But, you remember, for instance, in the case of Dr. Vickers' experiment, he had a clean spectrum, too. But, of course, he had a charge there. And then the answer to your other question is a matter of equilibrium. If you don't put the filter there, then you have a different case than if it's coming from the outside.

MR. PHILLIPS: Right. But why would you expect to see twice the amount of C prime as you did A, irrespective of the equilibrium? I can't explain that.

MR. GEORGE: You said you have no filter in the cup?

MR. PHILLIPS: Correct.

MR. GEORGE: So you're getting the radium C prime from the decay of radium A that's in the clean cup, and if you remove the filter you get an additional radium C that's in the air.

UNIDENTIFIED SPEAKER: But you always have more A than you have C prime in the air.

MR. GEORGE: I don't know much about spectrometers, but I think that's probably one of the reasons.

UNIDENTIFIED SPEAKER: How do you get more C prime than A in the air?

MR. PHILLIPS: I would believe, though, that if you had the filter on the cup and you left it for a sufficient time, wouldn't you expect a hundred percent equilibrium in the cup? That's what I would expect.

MR. GEORGE: I don't know.

MR. PHILLIPS: So I would think that the highest count would be when the filter was on the cup. I think that you'd have an equilibrium situation.

MR. GINGRICH: Excuse me, Chick. But I'm pretty sure you wouldn't expect a hundred percent equilibrium in that cup. The reason for it is that you've got a lot of plate out surfaces.

MR. PHILLIPS: Yes, but what I'm looking at is all the plate out. Because I'm seeing a clean spectrum. I'm seeing nothing from the walls. Or, essentially nothing.

MR. GROER: Bob?

MR. HOLUB: One more. We can discuss this later in detail, but in experiments done by Eugene Benson at the University of San Francisco they had a cup, a collimator, and it was supposed to measure radon daughters in the air together with radon by means of collimating in such a way that from one volume you would get only radium A and from the other radium C prime and radon. In other words, a perfect system. However, the collimator--it's really a sink for radon daughters for plate out--is just a few centimeters long in diameter, quite small. And still the daughters practically never made it, the alpha particles from those appropriate volumes never made it to the surface, to the track etch. If you look at the depletion of concentration of the daughters close to the wall, it simply is that there are no radon daughters. It happens very quickly and it must be very complicated inside the cup. You simply can have practically no daughters there. It can go anywhere almost. I'm not surprised that you see all kinds of things.

MR. PHILLIPS: Well, I would agree wigh you, except the only plate out I'm looking at is that which is on the detector. I'm not seeing the plate out from the walls.

MR. HOLUB: That's right. You are not taking care of the whole thing. Maybe it's escaping here and there. Almost anything can happen.

MR. PHILLIPS: Well, you might be right, but I don't know why taking the filter off of the cup makes it behave that strangely.

MR. HOLUB: These cups, when you touch them they're very easily affected--four thousand volts, and then inside it's plus and outside it's minus and there are spots that are even higher. So it can happen just by touching it.

MR. PHILLIPS: Yes, but it's the same every time, and I don't think I touched it.

MR. HOLUB: No, no. You have ten cups altogether and you take the one apart from the other and they are all completely different.

MR. PHILLIPS: I realize that. But every time I performed the experiment I just described, the same thing happened.

UNIDENTIFIED SPEAKER: Got the same results regardless of the variance.

MR. HOLUB: Then I don't know.

(Laughter.)

MR. GROER: The record should show that. I think without a blackboard, it's sort of hard to follow. I've sort of lost track visually at some point in this discussion. It's a very interesting question, but I don't think we can settle it. At least, my visual imagination is not good enough. Maybe with a pad of papers it can be settled at dinner this evening, I don't know.

Time passes quickly. I've asked in the initial phase for input from the audience. I would now turn the discussion around so the speakers can ask our panelists to make comments, suggestions, raise questions. I will start on the right end of the table here. Ron?

MR. COLLÉ: I think I've said enough. I have nothing else.

MR. GROER: Okay.

MR. WICKE: I want to ask Jim about his experience with charging his cups, if he did some experiments on this. It seems to me an important factor because when you make calibrations you assume that the distribution of the daughters which are plated out in these cups is reproducible. I think it might be due to charges that it is not reproducible, and that will produce some errors.

MR. GINGRICH: I think the whole question of charge on the cups has been overplayed a little bit and I say that because of our experience of making a lot of measurements, not that we've made a lot of precise charge measurements, but a lot of radon measurements. We've calibrated these detectors in something like six or seven different chambers over various periods of time under quite different conditions and so forth, and yet the data all fall pretty good on that calibration curve line. If there were charges that are maintained on the cups for long periods of time, and other considerations like that, I'm sure that we would see a much bigger scatter in our data. And we just do not see that scatter. So that's one empirical reason we think that the charge thing is maybe overplayed a little bit.

MR. PRICHARD: I'd like to direct some questions to anyone who may have been doing some work with activated carbon or other sorbents in concentrating or making integrated radon measurements. In particular, I'm interested in relative humidity and volume dependences. Any comments? Andy?

MR. GEORGE: Now, as I said earlier this morning, we just really began to look into this M-11 activator charcoal canister, and we haven't done too much in terms of humidity effects and temperature. Although I looked at the temperature range that you find normally in the indoor environment--and I assume for this kind of application for getting data like screen data, I thought that was sufficient. As I said, this area needs to be investigated. So I don't want to say this technique is ideal until you check it further. I'm only suggesting that it may be a convenient technique, and it's very easy to test this technique.

Now Jess Thomas back many years ago did some of these experiments, and I know there is some application, and I thought I sent you a copy. See, I don't forget. I sent you a copy, and I thought maybe you found the information that you needed. If you are not happy, I guess you have to contact Jess Thomas, because he spent a lot of time on that.

MR. PRICHARD: I was referring less to the cases in which you actually pull air through the canister. Where you just use the canister, rather, a volume of charcoal as a passive absorber, such as the previously mentioned unit and the M-11.

MR. PACER: We did some time tests using charcoal as a flow-through device. At room temperature and humidity, which in Grand Junction is probably about 10, 15 percent (we just wanted to get some initial numbers) one M-11 had absorbed something like 90 percent of the radon that it was exposed to within the first five minutes, and then it absorbed up to 97 percent after the first hour. The other units were a lot slower. It took about 20 minutes to absorb the first 85, 90 percent, and it took on the order of four hours to get to 97, 98 percent. So, there is a time dependency on that absorption, and I think it has to do with the difference in the volumes of the charcoal.

MS. HINKINS: I just have a couple of comments. Since I'm a statistician, my comments are mostly based on the data. This is probably not a discussion topic and maybe it's something you all do already, but it seems important to document your data in the sense that if you want to go back and look at the calibration results you can find not just the amount of time that the devices were in the chamber, but the actual dates they were in the chamber--not just the total exposure, but the radon concentration and the exposure time. And as much data as you can think of to put in. That's very helpful when it comes down to analyzing the data.

And one other comment on calibration. I find the method of displaying the calibration data a little confusing. From my background I look at it as an independent variable and a dependent variable and you're trying to fit a slope. And, for a Poisson distribution, a better estimate than the sum of the ratios is to take the ratio of the sums. Those are the only comments I have.

MR. PACER: I just have two comments. One is, I agree that we need some radon standards and I think we'd support NBS doing that. The other thing is, one of the things I was interested in coming to the meeting for was to learn something about radon flux measurements and their calibration, especially since EPA does have preliminary standards on radon flux. And I really haven't heard anything at all about that.

MR. RATHBUN: I would just say that when I measure radon flux, I do it with an M-11, and when I want them calibrated I just send them to Andy George.

(Laughter.)

MR. GEORGE: I guess you want to put me on the spot again. Well, we have been doing radon flux on and off, but we really don't do it on a regular basis. We tried both the accumulation method and the radon flux canister: Both methods work equally well. If you want data in a hurry, you do the accumulation method because you can do it on the spot. All you need is a can you turn upside down. You invert it over the soil with a couple of vials and let it emanate for maybe a half hour, no longer than two hours or you get into problems. You start building the radon in the can and then start pushing that into the soil, but you have to be careful. In our experiments with this, we found that for different size cans the time that you can allow for this build-up varies. So, we usually limit it to two hours. From soils, two hours is sufficient to accumulate enough radon, then you can transfer it to a Lucas-type flask and count in the laboratory, and then after that release the flux.

Now, on the other hand, the canister is a very convenient tool, but you need a much longer time for accumulation, usually, on the soils, something on the order of 24 hours. From the surfaces and the concrete basements or the walls, you need approximately 48 to 60 hours. In both cases it works fairly well, and canisters can be shipped back and forth very easily. In our experiments, we found that if you seal them after you take them apart from an illumination area you do not lose any radon. We counted them immediately after we moved them and then we sealed them and counted them over several days after we closed them up. We found no loss of radon. The reduction you see is sort of a decay of radon. So, they both work well, and you can measure almost any surface with these two techniques. There is no limit with this. Sensitivity, as I said, with the charcoal canister is about two-tenths to the minus eighteen curies per centimeter squared per second. I don't know what EPA is using as a standard, but I think one of the numbers they mentioned was like two hundred times the minus eighteen curies per centimeter square per second, after you clean up the tailings. Now, we found numbers higher than that in soils in the United States. That's background soils. They can go up as far as 500, with averages about 200 to 300 over an entire year. So, I don't know whether that is above background or is just 200; I don't know what the standard is. Does anybody know the answer to that?

MR. BERNHARDT: In response to your question, the proposed number right now is two picocuries per meter squared per second. It is within the observed background values, yes. I think it's intended to be excess above that.

Could I just make a couple of comments on what we have done on flux measurements? I don't have a lot to add to what Andy George says. The main thing we have used in Las Vegas is the accumulation technique. I guess I'll note that when you're applying it more to a research thing or trying to put more effort in I like the concept of taking several accumulation points so you plot a slope of the accumulation in the vessel and you get a slope-type thing versus just a single accumulation point.

We've done a little bit with the charcoal cartridges to get comparison. The comparisons have been reasonable. We really haven't pursued it as far as we would like. I guess agreeing with your concept of a problem, the accumulation vessel is only good for periods of like an hour to two hours at the most. It is hard even, therefore, to compare that to a charcoal cartridge. I've compared them over the same periods of time, but you're pushing the sensitivity of the charcoal cartridge by trying to only do the charcoal for several hours. It's difficult to do an accurate accumulation over the one week period that you need for the charcoal. So, to get a direct comparison is a bit difficult, but it's not impossible.

MR. GEORGE: Now, as far as comparing these two methods, we do have a radioactive concrete, with which you can do this kind of an experiment. It's a very homogeneous mixture, and it's big enough to use flux cans and canisters, and then you can remove the canisters. You can take your samples with the can and then remove the canisters at different times, and you can determine this kind of a relationship between the two. I agree with you; it's very difficult. In the field one time we had to stay overnight to remove canisters and take the flux can measurements, taking a measurement every two hours. You have to identify a whole area and place different cans every so many hours. That's how we found out that you cannot exceed, let's say, in this area, five hours. You start leveling off and you're getting out of linearity.

MR. COLLÉ: An alternative to sending it to Andy that seems to us to be quite feasible would be to actually develop the relatively small artifact standard for radon flux, which would be a physical surface that would not be too large, that literally could be sent to various laboratories for their own standard. We've looked into this in a preliminary way, and, although a lot of development work has to be done, it seems that it is quite feasible and that it would have a dynamic range over which it would work and that one could literally dope this. It would be a ceramic material of perhaps a meter by a meter. Thickness is somewhat indeterminable, but it would be impermeable on five sides. The exhaling surface is only on one side and you would only be allowed to use an inner portion of the surface area. It would be suitable for charcoal canisters or for small accumulators; and it would have a dynamic range under which it would work.

We have considered this quite extensively. If someone would like to talk to us sometime about perhaps picking it up, developing it on your own, we'd be quite willing to talk to you about it. It does seem like it's feasible.

MR. ROESSLER: I'm surprised we haven't heard from either Chick or Sam Windham about this whole question of flux. It's true that both the accumulators and the charcoal cartridges do a pretty good job of measuring the flux that occurred during the time interval for which the device was deployed, or if you're doing successive sampling you can sample a week and get a good measurement for a week. But the real question is whether this is a useful number for us because of the temporal variation that by hour, by day, by week, whatever, seasonal variations. Usually, the meaningful parameter for evaluating, in our case, a piece of reclaimed phosphate land or some other situation is the long-term average. And we can't think of anything short of a long-term sampling experiment to tell us what the long-term average is. So that's the real problem as far as I can see. What is a measurement that tells us what the long-term average flux is? These sorts of measurements don't seem to be that unless you repeat them over long time intervals. The question probably is, then, is there some other parameter that's more stable with time?

MR. KEEFE: One of the problems that has perplexed me for quite a while is the problem of plate out, which I've mentioned before. The most disturbing part of it is the inconsistency of the various different answers I get from different laboratories and different people. It seems as though there are a thousand different answers as to what effect humidity, temperature, charge and so on have on plate out. I think that's an area that needs to be tied down before we can develop good instruments that will accurately measure working levels of radon daughters.

MR. GEORGE: I wanted to say something about plate out in the design of instrumentation. Our experience so far shows that plate out is only related to the number of particles in the air. Our experiment (we have a paper on it in Health Physics coming out soon) shows that if you can measure the particle concentration in the room, you can almost predict plate out exactly from theory. All you have to do to test if there is any plate out effect with your instrumentation is to take your instrument, put it in a calibration chamber, generate a known particle concentration and, if possible, a known particle size, and then repeat this experiment with your instrument at different concentrations and go from high to low. If you see any reduction in your measurement, let's say of working level, then you know you have plate out. If you have no reduction, then you can assume you have no plate out. I suggest that it is one way of testing for plate out effects. As a matter of fact, every time we design an instrument we go down to five thousand particles per CC because I don't think you're going to find many atmospheres with particle concentration less than five thousand particles per CC. So I consider that a lower limit of particle concentration to which you should subject your instrument. I don't know whether you can do that in the mine because you have to have a filter to make sure that if you want to test for low particles you have to filter the air and perhaps inject some known aerosol concentration so you can maintain your chamber at the same level for some time until you get reasonable results.

In a chamber this can be done very easily. You can put in an absolute filter and then you can inject your aerosol, and you can control it very easily. Particle concentrations from five thousand to a hundred thousand are very typical. And I think most of our data show that you begin to get some plate out in some instruments that we've seen around between five and ten thousand particles per CC. About ten thousand we couldn't see plate out in any instrument. So if you can do that, I think it's a beginning. If you can check it around five and ten thousand and you don't get a plate out effect, you can almost forget about it. But if you want to be really scientific, go down to five thousand or maybe three thousand. Then you can argue with everybody.

MR. GROER: Jim Gingrich.

MR. GINGRICH: I thought it might be interesting to set something in perspective. You all have read some of the Three Mile Island accident analyses and so forth that indicate a very low potential number of deaths from the Three Mile Island accident. Right now, based on very limited data--and it's obviously very limited--on eastern Pennsylvania, we have about 15 percent of the homes giving radon values above 20 picocuries per liter. If you convert that to working levels (again, above .1 working level) and use the same criteria that were used for the number of people in the area around Three Mile Island and assume that 15 percent of the homes have radon levels of this magnitude, you can see that the potential lung cancer deaths are rather large.

MR. GROER: Thank you very much, Jim. It is a very interesting slide, but I feel very strongly we should stay out of the area of epidemiology. There are very special meetings for this sort of thing. Please don't take me wrong, but I think we should stay out of epidemiology. There's a meeting, as a matter of fact, coming up in October in Golden. Many of you know that. So there will be some discussions on this very topic.

Every one of our panelists has had a chance to comment. I'm coming back again to the audience. Any hot items you would like to see discussed, questions?

MR. TOOHEY: This isn't a very hot item or question, but it's something that you might be interested in. And that is, if you want to take grab samples of radon in houses, how do you do it? Normally what we've done in the past is we take a Lucas cell out and have somebody bring it back to the lab and count it. Well, that's nice if you're doing a hundred or so houses as we've done before. But in the next step in our study of ambient radon levels in Chicago area houses, we want to do between three and five thousand; namely, by giving everybody who works at Argonne a sampling device to take home, take an air sample and bring it back in. So we need something that's fairly rugged, will hold a vacuum, and is also cheap. And what we have come up with are these things, which you can pass around if you want to look at them.

They're basically aerosol paint cans, and they're 80 cents apiece. I don't know exactly what their volume is. My guess is it's about three-tenths of a liter or something. We get them from a factory; they say they are evacuated. We've measured that they only get them down to about 40 millibars. So we pump them down to almost zero. We take them into a house, give them to somebody, and just tell them, "Hold it in the middle of the living room, press the button." Instead of something coming out like it does in your normal aerosol can, something goes in. We then take them back to the lab, hook it up to an evacuated Lucas flask, let the two equilibrate, and then count the Lucas flask as normally.

This week is our first real test of these things under more or less field conditions. We filled four of them yesterday over at Ed's chamber at EERF. He's going to fill another four of them tomorrow for me. I'll take them back with me and we'll count them next week and I'll let you know how well they worked.

UNIDENTIFIED SPEAKER: How do you hook them up?

MR. TOOHEY: To what, the Lucas? The cylinder? Okay. Well, I'm glad you asked that question, Senator. We have another transfer device. This end will simply connect to the Lucas chamber somehow, with a shorter piece of pipe, of course. I brought this one down to hook up to the chamber. On the other end there's a piece of brass tubing inside a little Tygon sleeve. We take the button off of them. This just fits over the needle valve on here, you depress that, and it will hold it down and they equilibrate that way. We did these on a sampling port at the chamber, just by hooking the tube up and then slipping them over and pushing it down. We are making the assumption, which we'll test when we count them, that the valve wasn't depressed until it was actually in contact with the radon atmosphere.

But, as you probably know, Lucas cells are kind of expensive. They're fairly sturdy but not all that sturdy, especially around the windows; and they don't hold a vacuum for more than a couple of days. So we hope this will solve some of our problem with it. Maybe by the next time any of us get together I'll have some data on just how well they work. And maybe we'll put a note on something fairly brief on it in Health Physics or something. But we think this is one way we're going to have of doing a lot of grab sampling fairly conveniently.

MR. COLLÉ: I just have a question, and it's something that we have been burned on ourselves relatively recently. I seriously question when you hook up this can to the cell whether you're indeed going to get first mixing and then any type of equilibration. I really wonder if that's going to work. Before you go out making five thousand, I suggest that you check that transfer, because those transfers are more difficult than they appear on first appearances.

MR. TOOHEY: On, I agree completely. That's one of the reasons I brought them down this week. A twofold thing. First of all, see how well they hold up under field conditions. We presumably will know the concentration in the chamber. Sue Fisenne also took some samples in the bomb-type devices and some other samples have been taken. So we'll have a pretty good handle on what the concentration was and then we'll see what we get. So, maybe they will work; maybe they won't.

Certainly we will validate the technique before we go into the sampling. But that's another year down the pike probably.

MR. PRICHARD: Is there some particular reason you chose to go this route rather than, say, traditional plastic bag of some size which could then be blown up and returned to the laboratory for, well, possibly even concentrated techniques? Here it looks like your effective sample size is going to be somewhat less than 100 ML, accounting for the partial pressure.

MR. TOOHEY: That's probably true. The primary reason is to do it extremely simply. We don't have to send somebody out with these things to take the sample. It's a difficult question of what it is you want to do. Granted, grab sampling has its limitations. We all know about diurnal variations, this, that, and the other thing. Arthur Scott will tell us tomorrow maybe how to use grab sampling to decide what to do.

But we thought the first thing we should do is just get at least, one, preferably several air samples out of a lot of homes and see what we've got out there--because we've been surprised by what we've seen so far in a few homes. The highest we've seen is about 60 picocuries per liter. But, 10, 20, 30, is becoming kind of common.

This is a first stage of the project. Then we want to look at this and decide how to do a more accurate measurement if need be, or more houses if need be. And look at some other things. What really determined the level of the house? We're tending right now to think it's the geological characteristics of the soil underneath it.

MR. PRICHARD: Well, I agree with your goal of doing some preliminary grab samples. I may be missing something, but I don't see what's so difficult about sending people home with a bag of some sort to blow up, especially when you consider your potential problems with loss of vacuum or transfer problems.

MR. TOOHEY Well, I'm not that familiar with the bag technique, so I can't really comment on it. But it seems to me that there would be a lot more problems there than handing somebody an aerosol can, which he's already familiar with, whether it was paint or bug spray or whatever, and saying, "Just hold it up and push the button."

MR. COHEN: You have a volume 95 aquarium pump and a $12 bag--have them taped together--and tell them to put the plug in the wall and leave it there for five minutes and pull the plug out of the wall and that's the end.

MR. AKERBLOM: I would go back to a topic that we discussed earlier, the reliability of instruments. When you buy different field instruments--track etch, meters for measuring, you are rarely told how they react to humidity, temperature, and so on. I think that we should go together and actually tell the producers of the equipment that if they sell us the equipment they have to tell us much more. We have found it very difficult to get such information from producers. We tried in South Africa to get some figures on the Rowak caps. They haven't given them yet. They say they work. Okay. They work. But how?

MR. GROER: Okay. Any manufacturers here who want to comment on this?

(Laughter.)

MR. COLLÉ: I think the presumption was made that the manufacturer knows and is not telling you. In fact, the manufacturer does not know. I mean, most of the instruments that are manufactured were usually developed by someone like EML, in which case they got the specifications, mass produced them, wrote up a sheet for it and a price list, and distributed them. And that's the extent of what they know about them.

MR. AKERBLOM: Yes, that is perfectly true. They don't know. But they don't seem to test it either. They should have their instruments tested before they sell them. I found this with track etch. We had to start in Sweden to test the track etch. The manufacturing company should know if it was good enough. If we do not put pressure on the manufacturers, we never have any good instruments.

MR. GINGRICH: I guess one of the comments from our end of the line is that we welcome questions of any kind. Quite often people will ask us questions after they've been using our services for a while, not knowing that we may have already solved that problem for somebody else. So if you've got questions about our devices or how to use them, we would more than welcome any kind of communication and questions that may come up. There are still a lot of things we don't know and would be happy to admit it.

MR. GROER: Silence? I'd like to bring the discussion then to one question that has arisen for me. All the methods assume, among other things, that once a radon daughter atom or aerosol particle attaches to the filter that it really stays there. Is that really so? What do you people here think?

I'm referring especially to a paper by Johanson. Some people have probably read this paper. And I was really soliciting comments on this paper. Bob?

MR. HOLUB: I think he concludes that nothing really gets out. If it does, it's in a vacuum.

MR. GROER: Yes, that's one point. This paper refers to a study in vacuum. Now, my experience has been that in air I have no evidence that daughters are leaving without being asked. So, once they're in the filter they stay there. I have a very poor understanding of the mechanism by which filters work. I'm thinking of membrane filters. I thought that it might be worthwhile to get into this if there's interest.

MR. HOLUB: The University of Illinois, similar to Dr. Postadorf's group in Germany, is now examining the possibility of reentry from rough surfaces, whether it's grass or rocks sticking from the wall in a mine, into the airstream after recall. So it's definitely not very serious, and it's related to this problem. But the assumption for these studies is that there must be an airflow because other people already found out that if you have just recall then it simply returns to the surface where it was before.

MR. GROER: Okay. Well, there seems to be then a universal agreement, in the absence of any other comments, that daughters do not leave the filter. Okay. Then, maybe I can go on to the next assumption that concerns me--constancy. Now, I'm turning directly to Bob. I know that some years ago, three or four years ago, you started an investigation into time dependent concentration during the time of sampling. In one of the papers I wrote it said, "Bob Holub, private communication." Have you followed this subject? Where does the matter stand?

MR. HOLUB: I'm very sorry. It's still lying on the desk and nothing happened.

MR. GROER: Okay.

MR. SCOTT: Some years ago I did a study on atmospheric radioactivity, and this was piggybacked onto an atmospheric dust collection thing. The results then were somewhat disappointing because these measurements, because they're collecting dust, have to take place over about an hour at least. There were evidently changes in concentration there, and, despite analyzing everything with a multi-Z-squares fit program, we got nonsense answers out of it. This was particularly true if the radium A concentration increased near the end of sampling, because you then got radium A being one quarter of the concentration of atmospheric radium C or B. So, it's hard to say how large an effect it may have, but certainly if it does vary, it does make a large effect.

MR. GROER: Okay. Thank you very much. It is obvious that to evaluate this really quantitatively, by measurement, is impossible, because all of the methods you have assume constancy, and, if you want to check constancy, which methods are you going to use to do that? So, it has to be done indirectly. And I have seen instances where this assumption was certainly not true. One way of checking is to count while you are sampling. In this case we put the detector--it was a surface detector--underneath the filter paper. If the concentration is constant, then the radium A count you observe, say, during the first minute of a two-minute sampling to the count to observe in the second minute should be a certain ratio, within, of course, statistical fluctuations. If that ratio is not what it should be, assuming you take proper care of the statistics, then I think you have an indication that this assumption is not true. So that's what we have done in this area.

So what this means to me, and I think you will agree with that, is that there should be a trend to go to smaller instead of larger sampling times when you want to measure low concentrations. If you agree with that, I'll go on to something else. Okay.

MR. COHEN: This is a practical question. If the detector is underneath the filter paper, what supports the filter paper? The filter, I mean.

MR. GROER: If the flow rate is not too high, sometimes you don't need a support for this. Filter paper is really a plastic material. Now, sometimes you can support it with a grade of a certain reasonable mesh size. So, you lose a little bit of detection efficiency, but it doesn't really make much difference.

MR. KAHN: You could certainly test this, if you had two collectors, by offsetting the collection by a minute or two. See what happens then.

MR. GROER: Good point. I think maybe I don't quite understand you, but the problem there would be, can the two collectors be exactly at the same place in space?

MR. KAHN: Obviously not.

MR. GROER: Right.

MR. KAHN: As a matter of fact, if they were in the same place or pretty close to the same place, they may tend to interfere with each other regarding the air pull. But, if they did seem to be consistently similar, even though they were a couple of meters apart, that would reassure you.

MR. GROER: I agree with that. We've tried to do it in one place, and that's the only way I could think of looking at the ratio. Any more comments on that?

MR. NAZAROFF: It seems to me that there are two possible time trends that you would be interested in. In one case, if the fluctuations are of a fairly high frequency compared to the period in which you're measuring and there is no long frequency bias (I can't say this with total assurance, but it's my impression), it won't make very much difference in your analysis procedure. That is, if you collect a lot of radium A now and don't collect any for a second interval, then a lot and then a little and then a lot, and there are many of these intervals during your collection period, you are getting to the point of talking about discrete atoms--if you take it down to fine enough time periods.

The second sort of case where you would get into trouble is when the period is of the same order or greater than your sampling period. Some evidence can be gained in that question by making rapid successive measurements of radon daughters at one place. If you do that and see big differences over a fairly short period of time, then you begin to suspect that maybe this is going to be a significant problem.

I've done this sort of thing in one house, but the measurements were only made at 40 minute intervals. I could see trends of longer frequency than that, and they were not sufficiently rapid to indicate that there would be a problem with it. I guess that doesn't conclude that there couldn't have been things going on in shorter time intervals that could have caused the problem.

MR. SCOTT: Maybe I could comment on this question of fluctuation in time periods. The important question really is, "Under what circumstances are you measuring?" If you're measuring outside, then we would know what the frequency distribution of fluctuations in the atmosphere is. If you're measuring inside, we have a totally different spectrum of frequency--a totally different spectrum of frequency variation.

Now, given that our main interests, and I'll make an assumption here that we are measuring inside, we know that we have an effective filtering mechanism which cuts out the shortest periods of all. So the real question is, "Are our sample times sufficiently short that we do not expect there to be a significant variation?" We have done measurements with the Nork gas tracers, with spot samples as short as 30 second intervals; and you can show that in a house there are indeed fluctuations of the order of 30 seconds. But, in fact, the major fluctuations are of a considerably longer period than that.

A continuous radon monitor run so as to give repeated spot samples demonstrates that in a house there can be significant changes of concentration in a period of a half hour. In a room with a radon source in it the radon can change by a considerable number of percent in as short a period as five minutes. Now, we do have an advantage by measuring daughters in that things are averaged to some extent because the daughters we measure now are not due to the radon that is in the room now but due to the radon that was in the room ten minutes ago, 20 minutes ago, 30 minutes ago.

So, I suspect that if you don't put your sampling period much over about five minutes--and, in fact, I would raise another question, Why would anyone measure for more than five minutes?--that is quite adequate sensitivity. The problem is not likely to be of great significance indoors.

MR. GROER: Did you say the problem will not be of great significance indoors?

MR. SCOTT: I think so.

MR. KEEFE: In some of the measurements that we've made inside the home at Argonne, you can definitely see the effects from an air conditioner fan turning on. (We take measurements every ten minutes or so. We take grab samples with a nine minute measuring cycle, with a three minute pumping time or sampling time every ten minutes.) Up goes the radium A concentration and the equilibrium shifts. I suspect you can see as much just by somebody opening the door, disturbing the equilibrium in a home. If you have these instruments in a home and you are making measurements, I suspect that you're going to see the effects of a change in the equilibrium concentration of the radon daughters during that three minute time.

MR. HOLUB: What we propose is simply to simulate it--make a step function first, of the sampling and then nothing, and do it numerically. The Canadians, the group in Toronto, did something like that last year. Of course, it makes a difference, increases the error of all these methods. I forgot exactly how much.

MR. GROER: Thank you. I think those of us who are inclined to modeling and playing around with the numbers should really try to get at least a feeling for what happens if the concentration changes during the time of sampling. There were some experiments, but, as far as I know, maybe with the exception of this Canadian attempt, nobody has tried to get a quantitative feeling via modeling this effect. Maybe I'm wrong.

MR. SCOTT: As a matter of fact, I'm working on that now.

MR. GROER: Thank you. Any more comments on this matter?

MR. PRICHARD: While certainly not ignoring some of the interesting computational problems that would be introduced by changes in the daughter concentrations or equilibriums while you are sampling, I don't think I'd be in any danger of violating your suggestion that we not get into epidemiology by pointing out that the real bottom line in all the measurements we're taking does relate to long-term averages. You know, lifetime exposures to people. The end thrust of all our computations should be those measurements which help us determine those life-long averages. While there are many interesting and, in fact, fascinating technical problems, I would not want to see us lose sight of the forest on account of the trees, let alone the branches.

MR. GROER: Thank you, Howard, for this comment. Although I don't want to get into epidemiology, it is the working level months that are of concern. In the same vein, let me point out that there's another problem. The question is: Over what period of time--and after this I would like to leave this topic--should you integrate the exposure a miner gets? Should you integrate until the cancer was diagnosed, five years before the cancer was diagnosed, ten years before that? That's really a problem that has to do with carcinogenesis, and that's not solved at this moment. But I agree that epidemiology is based on accumulative measures. So there's no doubt about it.

Okay. Now, on epidemiology, any comments? If not, time is getting short. I would like to give the panelists one last opportunity to express their views, if they wish to do so. I will start again on this side of the table. Thank you very much.

(Whereupon, the meeting was adjourned at 5:20 p.m.)

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OPEN DISCUSSION II: AUGUST 28, 1981

Open Discussion: Measurements and Related Topics

Moderator: Arthur Scott

Panel: Speakers for Measurements and Related Topics

Richard E. Toohey
William W. Nazaroff
R. A. Washington
Charles T. Hess
Bernd Kahn
Carole Wilson
Gustav Akerblom
Andreas George
Stephen Rudnick

PROCEEDINGS

MR. PHILLIPS: The moderator for the discussion this afternoon is Arthur Scott. I'm sure Arthur by now needs no introduction, but I do have to tell you my favorite Arthur Scottism I heard him say once and I've thought a lot about during the last couple of days. He said, "The subtle things we see right away; it's the obvious that takes us so much time."

MR. SCOTT: Well, good afternoon and welcome. I see there are almost more people up here than there are in the audience. Nonetheless, I'm sure the panelists will do their best.

Now, the purpose of a moderator in nuclear physics is to slow down the neutrons so that they can interact with uranium, and I suspect the purpose of a moderator in a circumstance like this is to slow down ideas so that they can plummet into the fertile minds and imaginations of the members of the panel. There are some questions left over from this morning. So, it would seem to be a good idea if we started with questions. As moderator, I will take advantage of my position by starting off with a question statement, and I'd like to direct this to our Swedish colleagues who were speaking of introducing regulations for building.

Now, I happen to have had some experience with this, and I would be interested to hear what answers they have on two points. One of them is: What mechanism is to be set up to verify that the buildings indeed do meet a particular standard? Second: What is to happen if the building does not meet it? I'm dealing with new construction here. What mechanism is to be set up to check the houses?

MR. AKERBLOM: Well, there has been a long discussion within the Radon Commission in Sweden on how you should go on with this. What should be the standard and how should you actually tell people that they are below the standard limit or over the standard limit. We found out that there was no easy way to do measurements in houses and get the correct answer. You have also shown that today. You can't go in and take grab samples, and you can't actually rely on track etch films to give you a good answer, especially if you are measuring both radon daughters and radon at the same time.

We know enough to say, "This is regarded to be dangerous to people living in houses like this; something should be done about it." Should we be concerned about the exact number? Or could we say that in an area which exceeds approximately such-and-such amount of radon daughter concentrations, we would not allow people to live? In Sweden the amount considered uninhabitable is 400 Becquerels per cubic meter.

But whose definition of danger do we accept? It's not we who are measuring--we can only give our figures to the people who are working with it. We measure as well as we can. Should we be measuring more? It costs a lot. You do seven measurements the whole time, and still you wouldn't be sure, although much more sure than when you have done it. We have seen Scott's excellent work, yet he doesn't know everything. It's not low, it's not high.

No, you leave it to the health authorities. And tell them, "This is in a level where we think it's high; it's proved it's higher than it should be anyhow." And, if you measured over that level, you'd declare that place unsanitary. Once it is declared unsanitary, they have the means to go to the house owner and say, "This place is unsanitary." If he owns the house himself, he can do something about it. Perhaps he can just let it be. The laws are not so hard on that. But, if he's got children there, you can always start arguing with him about the risks for the children, and, in Sweden, where child advocacy is strong, it is not just his problem. The country also has to take into account the risks for the children. If he's renting the house to somebody else, of course, it's an easy thing. Because then he has to do something about it or he can't rent it out.

But this has been a discussion of people not willing to do something. And this will be difficult for a long time. Until now we have not met up with anyone who says, "I don't want to do this." "I don't want to follow your instructions." They are happy to know that you can do something to the houses. And you could tell them to enforce the ventilation in the house where concrete is used. But that would be a simple solution. But, if you were in a house where it comes from the ground, it's more difficult; it costs you quite a lot. So to give them a start, the State has provided loans for people who want to do something to their houses. The problem is, of course, that you don't actually know what to do. This is going to take some time. But we are coming out with some advisements which, I think, will be successful. They have been working in Canada, and I think they will also be working in Sweden.

Did I answer that question?

MR. SCOTT: Yes. Thank you. For the people who have just joined us, we are still beating the questions on this morning's session. If anyone does have any questions, would you please quest. Yes. The first one to the microphone gets it.

MR. RATHBUN: I just have a comment on the last presentation of the morning session, Mr. Washington's presentation. He was talking about the possibility of going to lithium fluoride dosimeters as opposed to the calcium flouride-dysprosium which he's now using. I would suggest that's not a good step since I had to go the other direction. You'll find that the calcium flouride-dysprosium has at least 30 times better sensitivity for gamma, which you're not really interested in, but it also has considerably better sensitivity for alpha, which you are interested in.

I used a .015 thickness, which is thinner than most TLD's that you get. Also, concerning the nonlinearity of the calibration factors, I also found this. But I found it to be much more pronounced at high levels than I did at low levels.

MR. KAHN: In connection with this, have you checked on the intrinsic beta background of the cans, just to see whether there would be a zero, non-zero reading?

MR. RATHBUN: Yes. Of course, I used a background dosimeter and always subtracted the difference out.

MR. KAHN: But they're not outside, are they? They wouldn't be exposed to the surface of the can.

MR. RATHBUN: They're not, of course, in the ionization chamber with the other chips, although I have run some right in the ionization chamber--but not on the probe. I found that the background measurement isn't much different above the case of the perm or below it or beside it. The place I had the biggest problem was unfortunately on one of our probes. It's designed to hold two background dosimeters on the side, and they are in a different plane, of course, 90 degrees out from the active dosimeters, so they don't get as much background if you stand them on edge like that. But, as long as you keep them flat, you're okay.

MR. KAHN: Well, I was just thinking that if there was a base background due to anything within the ionization chamber itself, that obviously would cause the curve to go, not to zero, but to the positive number.

MR. RATHBUN: Yes, I think if you use the background chip, you'll subtract out not only the gamma, but also the beta.

MR. GINGRICH: Back again to Mr. Washington's paper of this morning. I spent some time with the Australian Radiation Lab a couple months ago, and they had done a rather thorough electrostatic field evaluation of that particular device and came to the conclusion that most of the problem of the huge variability of the device was due to the placement of the TLD in the electrostatic field. They did not feel that that device would give them the kind of confidence that they wanted in making the measurements there. They had done quite a bit of testing on it.

MR. SCOTT: First one to the microphone gets it.

MR. PHILLIPS: This is to Richard Toohey. I've seen those slides before on the equilibrium values. Have you verified those values, using any different instrumentation than the two you described this morning? I think, namely, you are looking at radon with the Wrenn chamber and the working level with the Environmental Working Level Meter.

MR. TOOHEY: Yes and no. The Wrenn chambers are. We always take a few Lucas flask grab samples, and they seem to work pretty well. But we have not used another device to cross calibrate or check the working level monitor.

MR. PHILLIPS: Thank you.

MR. SCOTT: I'd just like to add a comment to this. First of all, the working level--equilibrium fraction ratio, rather--is defined as 100 times working level over a radon for a co-existing concentration. So, in fact, under normal circumstances, no parent measurements actually measure equilibrium fraction--because it is extremely rare for people to take air through a filter and put it into a plastic bag and then take the radon sample from the plastic bag.

So, we in fact are bedeviled in this question by a further source of variability. First of all, there's the spatial variability because you're not measuring the same space; and, secondly, the sample times are so far different. I mean, take a short sample of two minutes. It only takes a matter of a second or so to fill a Lucas flask. And, your sampling theory says that, your estimate must be different.

MR. TOOHEY: I agree completely. In fact, it's worse than that. We found that our particular Wrenn chamber takes about an hour to come to equilibrium with an external radon concentration. Most of the measurements I showed were hour-long counts. So, obviously, we're seeing an hour-long average, whereas the working level monitor takes a two or three minute sample and counts for ten minutes or whatever it is. This is certainly a source of some of the variability we see. But I think on a long-term average, hopefully, that will even out.

MR. PHILLIPS: I'm aware of all that. I guess the thing that disturbs me about some of that data is the extremely low radium A values when compared to the radon.

MR. TOOHEY: Well, most of it is on the surfaces. We have made the measurements, and we can show that the missing radium A has, in fact, plated out.

MR. PHILLIPS: I've just never observed that, I guess. If I make simultaneous radon and working level measurements in a room, I see a lot closer equilibrium of radium A to radon--in any situation.

MR. SCOTT: We had a number of papers this morning that touched on the topic of plate out generally. Is there anybody else who is interested in plate out or would like to say something about it? Mr. Holub?

MR. HOLUB: I have to say something about the fan, obviously. There was an additional up-date paper on plate out published in a proceedings from the University of Illinois in 1979. I can send a copy of the proceedings to anyone who's interested. The reason we did this was simply to try to understand better what is happening. The first thing one has to look at is the condensation nuclei concentration--its size, of course, its charge, humidity, and perhaps even the chemical character, of the condensation nuclei--and then try to guess the mechanism. So, for instance, it's much more likely that small condensation nuclei would stick to the fan rather than the big ones, which might bounce off because of inertia effects.

Then also there is the charge or even image charge on the blades. We have explained it in our paper--we had put charge on the blades and it affected the sticking. Then there is the big question of what are the sticking coefficients, because the typical aerosol phycisist always insists it's one. But there is now growing evidence from other sources--the Indians measured it, and certainly Hopkie measured it, and Krugar in South Africa--that the sticking coefficient is much less than one.

All this says that one should be careful about assuming what's on the fan and what's on the wall. It depends on many things. So I would somewhat take exception to calling it an artifact. It's the most natural thing.

MR. SCOTT: And what is probably a concluding comment on this, I've been looking at this question of deposition, too, by measuring deposited activity on the walls of buildings. This is a field measurement, not a laboratory measurement. It is difficult because if one uses an ordinary alpha scintillation counter your sensitivity isn't that great. But, nonetheless, we do have a few places where the radon concentration is high, and we're able to demonstrate from this a little bit of theory that, in fact, deposition velocities for unattached daughters are in the region of .1 to .5 centimeters per second, which is what we measured, and will, in fact, account for most of the variation of working level ratios that we actually see in houses. So the question of squaring the material balance does not seem to be beyond the wit of man at the moment. The difficulty seems indeed that deposition velocity is a function of air turbulence--of air velocity over the surfaces. So, as usual, we were able to replace one fundamental question by an even more fundamental one.

UNIDENTIFIED SPEAKER: I'd just like to make a comment. I have read your paper. You were nice enough to send it to me, and, as I recollect, I didn't refer to it in my write-up, but your deposition rates were about the same as what I was getting. So, I think there's some agreement.

MR. SCOTT: Are there any more questions arising out of this morning's presentations?

MR. TOOHEY: I'll follow in our esteemed chairman's footsteps by asking a question of a co-panelist, and that is Bill Nazaroff. In your paper this morning you talked about correlating the radon source term with wind speed measurements. I wanted to know what happens to the radon level. We've seen essentially no correlation between wind speed and the radon level in a house.

MR. NAZAROFF: For any of you who have been to a meeting with Arthur Scott before, if he was in this situation, he would pull out a view graph and show you exactly what had happened, but I'm unfortunately not that well prepared.

We made wind speed measurements in only one house. When you look at the radon data from that house, you see that when the wind speed increases, the radon concentration increases slightly and then after a few hours begins to drop in response to the increased air exchange rate. You could, in principle, take the figure that I presented and work backwards, but I have the original data which I would be happy to send you.

MR. GINGRICH: I just want to make a comment about the wind speed thing as long as we're on that subject again. We made a lot of soil gas measurements with our track etch cup along with comparing our comparison measurements with some other techniques. One of the most interesting things we found, and it may have an application to Bill Nazaroff's problem, is that if you set out a grid of samples or sample sites in a very hilly area and make soil gas measurements, if you use a radon emanometer where you're taking grab samples, you'll find that when the wind is blowing onto the surface of this side of the hill the radon soil gas levels will be fairly low, in fact, quite low if the wind is high. If you then measure over the top of the hill on the opposite side, the radon levels go up again. This is due to either actual forcing of the air through the soil on the hill or possibly Bernoulli effects bringing a lower pressure zone on the other side.

The same thing may be true in these houses. When the wind is blowing on the house, it could be Bernoulli effects that are helping to bring radon out of the soil gas and into the house. Also, with regard to other measurements we've made in soil gas (we've made something like 300,000 soil gas measurements in different parts of the world), the average we're finding is about 100 picocuries per liter in the soil gas. But it's not unusual to have soil gas concentrations ten times this. And that's only 15 to 18 inches below the surface.

MR. SCOTT: I'd just like to reinforce this. If, in fact, the causation of high radon levels in a house is due to the pressure in the house being lower than outside, it's clear that Bernoulli's theorem requires that when the air blows around the houses, it's got further to go; the pressure inside the house must be lower. Therefore, the harder the wind blows, the more negative the house will become relative to some distant point on an infinite plane.

In addition to that, the upwind portion of the house is a positive pressure on it and the downwind portion of a house has a negative pressure on it; and these pressure zones do not terminate at the soil. They in fact extend into the earth to some distance. We have made measurements using an emanometer around houses of the radon concentration and soil gas and find that if you can make measurements on the upwind side, at times at a depth of two or three feet, you get essentially outside air. On the downwind side, you can get concentrations of perhaps a thousand picocuries per liter, which is normally the kind of concentration you expect at several feet depth.

The whole situation is incredibly dynamic. Many of the things are not. Many of the problems are partially aerodynamics, not those of the radium in soil, per se.

MR. COLLÉ: One thing puzzles me, I've seen an awful lot of results lately where people have looked at various correlations to meteorological parameters, but I don't see how you can find a correlation between radium content, or radon concentration, and a single variable unless you have some model you're starting with, since all of the meteorological variables are cross-correlated. How can you separate out that cross-correlation? I mean, temperature and pressure are certainly cross-correlated, you know, and wind speed. In other words, since seldom are all the other variables constant so that you can really see a correlation between radon and wind speed, I just don't see what these results mean.

MR. SCOTT: I have a particular bias on this subject. When we started on our remedial projects, everybody knew certain things were true. After a short period, I decided that the simplest way of evaluating them was to assume that everyone was wrong. And we made measurements in adjacent houses for a period of time and found that readings made within a matter of 15 minutes in adjacent houses were not correlated. Now, it is conceivable that for a house you could develop a set of correlations with wind speed and so on. If, for example, there is only one hole in the house for radon to come in out of the soil, when the pressure is high on that side then indeed radon may come in. But that particular correlation may only apply for that particular house. I would require a lot of convincing to believe that there is any general correlation with anything except perhaps the most physical of variables, such as perhaps wind speed, in the sense that may increase the Bernoulli forces. Though, unfortunately, it almost invariably leads to an increase in ventilation rates, so it's hard to see the difference, and perhaps the external temperature difference.

Presumably we've beaten this one more or less to death. Would anyone like to change the subject with questions?

MR. BAILEY: I have one general statement to make. I've seen a lot of times when people amass paired data and then start to do regression analyses, line fitting least square analyses. I think we need to go further than just the "R" factor. I think whoever is massaging the data needs to go look at the standard error of Y on $\bar{X}$. It's very easy to take a shotgun blast of data and find a straight line that fits it very well and assume that there's a good regression or a good correlation, when it's not necessarily so because the error of that analysis is very, very broad.

My second comment is directed to either Mr. Akerblom or Ms. Wilson concerning their limits for new structures of two picocuries per liter. In our study in the State of Florida, we found houses that are outside of our district of concern that we used for control structures to be in excess of two picocuries per liter. From that we assumed that this is sort of like background for maybe six picocuries per liter radon indoor, being background, or typical of anywhere. Have studies been done in that area on houses not presumed to be in the area of concern or houses built with crawl space or mobile homes or whatever, and, if so, what kinds of indoor radon levels are there?

MR. SCOTT: On the two picocuries per liter, I think that's two picocuries per liter equilibrium equivalent radon, or .02 working levels.

MR. BAILEY: Okay. I misunderstood the equilibrium.

MR. SCOTT: A good SI unit there, I think.

MR. AKERBLOM: It is really a problem. This is a very low limit, as I see it, for radon daughter concentrations inside your houses. When we had the problem in Sweden, it was decided, based perhaps upon too limited a number of measurements, that, for health reasons, you couldn't go with a higher dose and allow it in your houses. I think that is something one of you has to come up with--what dose will we permit for people living in houses? And, if you have not decided that, well, you have to do it sometime.

The Canadians have done it from what I understood from you, Mr. Scott, for houses on tailings. We have done it for houses on ground anywhere. But still you might say these are provisional limits, as I see it at least, which may not be generally applicable. We, in Sweden, had simply come to a point where we had to impose a limit. It was done before we had full knowledge.

MR. BERNHARDT: Just for clarity, is that net above background or is that a total gross, two picocuries per liter in equilibrium?

MR. AKERBLOM: It is a total measurement in the house. I think I misunderstood Mr. Scott's question earlier in the panel. He asked me how we measured this, and this, of course, is the question. Until now I don't think anybody has tried to measure this. There has also been discussion about how it should be measured. They say it's not possible with the equipment that we now have. So it is possible that one might permit a radon dose of double that amount before doing anything about it. However, in my opinion, anything over that amount should be considered an emergency situation requiring immediate attention.

MR. SCOTT: So, it looks as if we seem to have finished with questions of this morning's presentation. So I will, again, take advantage of my position and propose a few questions for the panel. It's always a good idea to start on a fundamental question, and it strikes me that in the group of people gathered here we have had people who are worried about making measurements in mines, people who are making measurements in the atmosphere, people who are making measurements in houses, and, indeed, there are people who are concerned about making measurements in laboratories. Many of these people, in fact, don't have much in common. For example, the question of plate out in sampling heads is a non-question for probably about 95 percent of all people who take samples because they use an open-faced filter holder and transfer the sample to a separate scaler. It's only the sophisticated guys who give themselves sophisticated problems.

But let's see if I can raise any interest on a fundamental question. We have an international meeting here on radon-radon progeny measurements. What's the purpose of the measurements? This is a fundamental question because it, in fact, raises a lot of questions and also simplifies things a lot. What purpose are the measurements that we're talking about? Is this a very general meeting or is there an underlying theme or interest that we should be pursuing?

MR. HESS: I think that people who measure radon should have a responsibility for measuring radon in their own area. I think fulfilling this responsibility can provide information from a wide variety of different sources of radon, and our picture then of radon levels in the United States or, for that matter, throughout the world will gradually become clearer.

I must say, I have a lot more enjoyment from measuring specific physical effects, but I think that if each of us could take it upon ourselves to measure some local buildings--either private dwellings or factories--and try to collect more information about what the levels are, we would then have a more complete picture that would help us determine some of these questions about levels.

In my case, I've spent most of my time measuring in Maine, and I have found that doing one State is more than enough trouble--more than enough difficulty to keep somebody busy for a long time. I think that as scientists in this area we have a certain moral responsibility to do some work which will be of significance, perhaps either for health or for understanding or to alleviate fears or misconceptions in our own areas.

MR. KAHN: In connection with that, there seems to be now three possible sources of elevated levels. One is in Maine--and some other places, too, I guess--where there seems to be a lot of water bringing in high levels of radon. Certainly in many parts of Sweden there seems to be the underlying geology of the shell bringing in radon. We've worried about building materials, but I haven't seen any that are as high as that. Even in Sweden it seems as if it's underlying material more than the building material. Probably a systematic way of getting at this is to say, "Well, okay, let's look at these two areas." Of course, there's also man-made materials being placed underneath houses. Let's look at these areas from the point of view of geology, for instance, for high radon water and high radon emanating materials. Then let's do extensive measurements. How about these places in Pennsylvania? What's the source there?

MR. GINGRICH: The area in eastern Pennsylvania we've been surveying is a known uraniferous area. There are some old uranium mines in the area and some underlying rocks that are known to contain uranium. It extends not only to eastern Pennsylvania, but down to New Jersey and a few other places in that area.

MR. KAHN: I'm sure in Maine, for example, you went at it because you knew there was a lot of radon coming out of water supplies. So, I think that if we wanted to go at this from a public health point of view, we'd need first the kind of thing that's being done in Sweden. I guess we can get at that one of these days in the United States. The EPA laboratory here has been looking at radon--how far are we with radon, Chick, in water? Will we within a few years know where all the high radon levels are in the water?

MR. PHILLIPS: I think we'll have a lot better idea. I think certainly within a couple of years we'll be able to identify most of the problem areas.

MR. KAHN: Okay, so we may have these four areas. I think a way to go at it is to not all go at it and measure but to define where these problems are and then look at the high level areas and the marginal areas.

MS. WILSON: I think, in fact, in the United States here you've got an awful lot of information that you haven't used yet. I've talked to John Pacer from Bendix and he says that large areas of the States have been known for prospecting purposes, and this information has not been used by the environmentalists. All our radiometric measurements were made for prospecting purposes. This was material that we had at hand and so we just decided to use it.

MR. BAILEY: We have had a commercial group do a radiometric survey of the north end of Florida for mineral mining purposes. In the southern end, the phosphate region of Florida, we did our own aerial survey for gamma only, just gross gamma exposure. There has been a lot of controversy going on between various groups as to whether or not gamma is a useful indicator of the radon problem or potential radon problem. We happen to think that gamma is actually of very little value whatsoever as far as a radon problem, unless the exposure is well over 25 or 30 microroentgens per hour. Otherwise, it's not much of an indicator at all as a potential radon exposure problem.

Part of the thing that we're stuck in the middle of--that probably many other groups are going to be in--is modeling. As in the case of the people from Sweden, they talk about two picocuries per liter being the maximum allowed in new residences. One of the things that we are looking for, that we're trying to do something about, is how can you go to a bare piece of property that you haven't seen before, except maybe driven past on the highway, and take a measurement, do a little handwaving and say, "Yes, we'll have a problem," or, "No, no problem here."? That's one of the problems that we're really stuck with in the States. It's not measuring it so much or what to do about it when you find it, because there's nothing you can do about it. You can't force anybody to spend money on their house. But what do you do with a piece of property with cows on it that a developer is going to build houses on? That's the problem that we're stuck in.

MR. RATHBUN: I have a question along that same line. I have sort of lost track of where we are in the scheme of modeling these days. I know that UDAD has gone through a certain amount of transition, and I hear people talking about AIRDOSE and MILLDOSE and I wonder how all these things are related and what the status is now.

MR. SCOTT: Offhand, it would seem that things like your MILLDOSE and AIRDOSE are not really particularly relevant to this problem. The difficulty is not radon coming in from outside, but radon from inside trying to get out. As to the question of modeling houses, I don't know how far that is along. Again, there are so many parameters that it's difficult to know where to start. This indeed is the unsolved mystery. So what impels the stuff to come in?

MR. BAILEY: This is sort of what I was trying to lead to with the question to Gustav and Carole. My second question was: Do you have a limit for a new structure? Do you allow the new house to be built and then go in and measure it and then say, "Well, you're past the limit, you're going to have to do something about it."? Or, are you developing a method for looking at a piece of property and saying, "Most likely remedial action will be necessary in this type of a structure."? If so, what parameters are you looking at as flag parameters to decide whether there should be modifications?

MR. AKERBLOM: In fact, we are looking for areas with high radon concentrations in the soil. It's not only high radon concentrations; it's how much air is available to get into a house. We need to have some way to tell if it's a big problem. In my opinion, all houses should be built a little bit off the ground surface. Even on normal ground radon comes in, as shown today from many of the reports on ground problems of radon in soil. You have to really, from the beginning, ask, "Are the building techniques what they should be? Are they proper to meet the requirements of health inside the houses?"

These concerns must be impressed upon the people who are building them. You have to implement some building regulations. Otherwise, we'll have problems elsewhere. We'll have it there and there and there.

MR. SCOTT: The problem to some extent is probabilistic. If you remember, I showed a view graph of the distribution of means in Elliot Lake. Now, this is an area where undoubtedly there is a radon problem by our definition of a radon problem. On the order of 20 percent of the houses were above .02 WL. Now, Elliot Lake is a relatively small and compact town. But, suppose we turn it around. We're saying in fact 80 percent of the houses don't have a problem. The soil, to all intents and purposes, is the same over this area. It's about as uniform as you're likely to get anywhere in that kind of area. It isn't like some areas such as down in Florida where the soil has been carefully resegregated, and it is probably even more uniform, though high. Eighty percent of the houses don't have a problem. Interestingly enough, we have some areas where there's an attempt to do radon proof structures, and 50 percent of the houses have the problem.

So the real difficulty is that there are some subtle effects taking place in the soil. We've even had different working levels, considerably different working levels, from one half of a duplex to another. If you cannot get uniformity over a distance of 20 feet in the parameters that control the entry of radon into houses, I think it is almost a vain attempt to try and talk of large areas except in the vaguest of terms.

If you could just paint the curbstone with zinc sulfide and find your way home at night, it might not be a good area to build on. But, even places that are essentially at the low end of the scale, less than one picocurie per gram radium, still have some houses in them that exceed .02 WL on the long-term average. So, one can only make probabilistic statements. It is well-known, now, that neither the public nor the regulators understand matters of probability.

We seem to have drifted off a little bit from the original question, which was: Is the underlying theme, or has the underlying theme of the meeting been, environmental-type measurements or measurements in houses, and should we, if we are thinking about the underlying theme, think about it more, perhaps? Suppose, in fact, we take for granted that really the main interest of the meeting has been matters of environmental homes. Then there are a couple of questions that spring naturally out of this, and one of them is: Are the current devices you've got good enough? I mean, should we, in fact, ostracize anyone who comes around for a grant to improve an instrument on the grounds that he's purely wasting the taxpayers' money? Or, should we, in fact, say that the present instrumentation is so inadequate that anyone who comes around proposing action on past measurements should be ostracized because he clearly can't determine anything useful? Or, indeed, is the situation in between the two?

Would anyone possibly on the user side like to say something about instrumentation and its present adequacy?

MR. HESS: I notice that McDonald Wrenn is not here and Henry Spitz is not here, so I'll say a few nasty words about Wrenn meters. They have a difficulty with humidity, which has been written about and published by Andy George and others. They also have the property that when you set them down vigorously some of the parts inside come undone. So you need to be somewhat experienced in repair of equipment in order to make field measurements with them. I would say that having a rugged electronic device, having one that's rugged enough to be able to be taken from one lab to another, ought to be one of the criteria applied before a device is suggested for common use in field studies; it ought to be able to travel around in an automobile and be bumped and so on. I guess that's not the usual degree of strength that you'll find in laboratory equipment at universities. So, "ruggedization," I think, is one of the things that I'd like to see in some of the equipment. Maybe that makes it more expensive, but I think it's well worth it.

The other thing that I'd like to encourage people to do is take meters from one lab to another and try to get multiple calibrations. I've found that very useful and also I learned a great deal by doing it with the Wrenn meter.

As far as the Track Etch cups go, I think I'm reasonably satisfied with the way they behave. I would really like to see, though, some type of a track etch device or something analogous to it that would be perhaps a factor of three or four cheaper. That would really open up surveys of a thousand houses, which would be quite nice. If something could be done to have a reliable detector that works like a track etch detector does but that would sell for perhaps $5.00 apiece or something like that, then that would be a very valuable contribution, I would say.

MR. PHILLIPS: I'd be interested in a good passive working level monitor--one that sells for under a thousand dollars and reads down to .005 working levels.

MR. SCOTT: As a user, I'd be interested in just getting a good reliable air sampling pump that was small enough and light enough to be carried around by a five foot two, 18-year-old girl, preferably with blond hair and blue eyes.

(Laughter.)

MR. BAILEY: In following what Arthur was just talking about, a year or two ago, Mr. Pai, up in the Canadian area, was working on a little, aquarium-like pump with a track etch intimately placed to a filter system. I got a letter or two about it, but has it gone anywhere, or has it gone into the fish tank?

MR. SCOTT: No, no. It's been saved from the fish tank. This is Lynn Pai of the Ontario Ministry of Labor, which is responsible for radiation monitoring. What he has is an adaptation of the French CAE nuclear track dosimeter for working levels. It isn't passive; it's active. That's why the pump is required. It uses cellulose nitrate film; and the energy sensitivity of cellulose nitrate is used in this in conjunction with the absorber so that you can, in fact, get the total number of radium A atoms, radium C prime, and also thorium C prime of that deposited on the filter. It's hoped that it will shortly be put into use in Elliot Lake, as a matter of fact, to monitor new construction for compliance with the new construction limit of .02 working levels.

MR. PRICHARD: While we're in the business of wishing for new and improved equipment, a little gadget I'd like to see on the market--please excuse me if it's there and I haven't seen it--is a simple and rugged and inexpensive pulse logger. I don't think we have so much a shortage of devices that are capable of translating an alpha particle into an electronic pulse, but, at least in the realm of field equipment what I have seen, there is a real shortage of handy little gadgets to take a pulse and store it until you're ready to ask it how many pulses it saw.

MR. SCOTT: Well, I was unable to provoke many flights of fantasy there. In the operational field, and, of course, most people here are not involved in quite that sense, we do have a continual problem that I touched on this morning, and that is how well the short-term measurements represent the long-term situation. Now, to get an annual average you must make measurements for a year. But one of the topics--if anyone has any views on it I'd like to hear it discussed--is the question of variability in the short run. You know, maybe a few measurements in the best of all possible worlds would be sufficient to give us an estimate of the long-term average. Can I stimulate any interest here?

No, clearly not. Well, we've heard one heart-felt cry regarding calibration and inter-laboratory comparisons. I think everyone who has participated in one has found it instructive, particularly if they came out in the middle.

(Laughter.)

Yes. Well, that's useful and instructive. It's instructive if you were one of the outliers. The general theme seems to be we can't have enough of them. Would anyone like to comment on those kinds of things? Are there sufficient numbers of these things? And what about the crude and untutored users of the majority of a lot of this equipment? I think, particularly, of people in mines. Is there sufficiently wide publicity given to intercomparison to persuade them to send in an air pump and scaler? Maybe one of the laboratory people might say something.

Yes, please.

MR. HOLUB: You mentioned pumps; it is really a bad problem. The best seems to be a self-regulating device. It costs about $300 and can control its flow to pressure up to 50 inches of water. We had the best experience with these. All of the others failed, and this one eventually failed, too, in the mine. But it was definitely the best. I can provide details on the pump to anyone who wishes them.

MR. SCOTT: Though, this doesn't quite answer my question.

MR. HOLUB: Oh, I'm sorry, I thought you asked about the pump.

MR. SCOTT: No, I'm saying, what about the user? It's all very well to have a good pump, but do the users ever get encouraged to actually send in a pump and scaler to compare the working levels of the climax mine with the working levels on your test mine?

MR. HOLUB: Definitely not, no. They don't really care.

MR. SCOTT: Indeed, a question of accuracy in a vacuum, so to speak.

MR. RATHBUN: I think some of this is done, although probably not enough of it, especially with the air pumps. I formerly worked for the Mine Enforcement and Safety Administration in Denver, which worked quite closely with the Bureau of Mines there and the people did send in those pumps and scalers--all sorts of equipment--to be calibrated by the government.

MR. HOLUB: I have to add that these recommendations are usually ignored. I am not complaining, just stating.

MR. SCOTT: The reality is, everyone checks. A given organization often checks its scaler with a source that's been around for five or seven years and has got someone's initials scratched in on the active side. Unfortunately, it so often seems there is a tremendous gap between what we know we can do and what we actually do. But, still, there seems to be no place to get self-confessions from the assembled multitude.

I see Andy George here sitting pensively, so I would like to direct a question to Andy. With all the instruments that come walking through his laboratory, when do you think we will have enough?

MR. GEORGE: I think we have enough already. It's just a matter of calibrating and using them properly. We have every conceivable instrument that you need to determine exposure, concentrations, and so on. So I don't understand why you need to develop any more. We just have to take what we have and try to do some quality control calibrations and start using them. I think our immediate goal is to go out and make a lot of measurements to find out what is the exposure throughout the United States. There are questions to be resolved. For example, Jim believes geological formations cause the high levels in Pennsylvania. I don't think so. We found radium concentrations in soils about 1.2 to 1.5 picocuries per gram. He can hardly say that's abnormal. So, there must be many, many other locations like that throughout the United States.

So as far as instrumentation, I think we should go out and take numerous samples and identify these areas in the country. We do have instrumentation, if we learn how to use it properly. All the instruments I work with are usually built in our laboratory and I can vouch for them. But I've seen a lot of instruments come in that are not working; and they are supposed to be working. They are off by factors of 2 to maybe 22, as much as that. So it depends on who builds the instruments and how much he puts into calibrating his instruments. As far as I know, no commercial devices have been calibrated at all. Sometimes they put a calibration factor from theory, but that doesn't work. For example, when you calibrate an instrument that uses a TLD dosimeter, you have to calibrate the whole system, including the TLD and everything. So, when you are using calcium fluoride or lithium fluoride, it is only as good as you can calibrate it. I mean the whole system--not just the PERM or the passive device or the active device--has to have the TLD included. Everything has to be included.

MR. PHILLIPS: I hate to disagree with Andy because he's certainly the expert. But I don't think we have a working level instrument with which we can make long-term measurements in a large number of homes without expending an exorbitant amount of manpower and money. Now, I think we can do it for radon, but, unfortunately, guides are not set in terms of radon. Even though we might all say that the equilibrium factor is .5, you still, at some point, have to measure the working level; and I don't think we have those instruments yet. At least, I'm not familiar with them.

MR. SCOTT: Would anyone in the audience like to comment on measuring the environment? Instead of talking, getting out there measuring? Oh, well here's a surprise, Mr. McGregor.

MR. MCGREGOR: It looks like Arthur is pointing in my direction. It's our agency that has perhaps made the most extensive use of grab sampling techniques--and I was quite interested to hear Arthur's comments regarding his synthetic grab sampling with his replica sampling procedures. To date we have measured in 19 communities now, and about thirteen and a half thousand homes. But they're all single measurements. We feel that this has given us some idea of what the general background levels might be in various communities and perhaps in various geological environments.

We have also done perhaps another five communities across the country to a much more limited basis of sampling. But it's rather interesting to hear Carole Wilson's comments regarding the work in Sweden using the geological maps that they have prepared for their aerial survey work. We have also noted in Canada some areas, Castle Guard, British Columbia particularly comes to mind, where there is a town down the Columbia River called Trail and there is a definite, significant difference in the average levels of radon and radon daughters in these two communities. Castle Guard is being affected by a large uranium potential ore body. It's about .6 percent U_3O_8 up the hillside which has undergone valley glaciation, at least twice, with the detritus being deposited in the valley and contributing to the higher levels in Trail--oh, sorry, in Castle Guard as compared with Trail. This has also been noted in geological differences in the Bancroft area for radon concentrations in homes and in northern Saskatchewan.

Speaking of Saskatchewan, there is a study currently underway with the provincial authorities using the extensive aerial survey maps to try to delineate high risk areas for radon in the community of northern Saskatchewan. They are using these maps and then going into the very small villages and towns and doing house surveys.

MR. SCOTT: Thank you very much. I think this shows that you can in fact determine quite a lot if you actually go out and measure, rather than sit around and think about how best to measure.

MR. AKERBLOM: While I have the opportunity, I want to ask if anyone has been doing radon measurements in underground homes, which seems to be a popular way of building homes in the States and is starting to be in Sweden. But, I think, if you have a problem on the earth's surface, digging yourself down into it will create a much greater problem. I would be very happy to get in contact with people who have done actual measurements in underground homes.

MR. TOOHEY: We have done some preliminary measurements; we've done six underground homes. I don't have the data with me, but I can easily send it to you. I do have the average with me, which is an arithmetic mean of 4.5 picocuries per liter for the six homes. We also have done some preliminary work on solar powered homes; specifically, a few which have essentially put a big rock somewhere in the home as a heat sink and reservoir. In those, we didn't see much out of the ordinary. But, on the average, the underground homes have been higher.

MR. SCOTT: Well, as we seem to have run out of inspiration, I think we will just conclude the meeting so that everyone can get a cup of coffee before they depart for the laboratory. I'd like to thank all our panelists and all you participants and turn over the meeting to Chick Phillips for the concluding remarks.

(Applause.)

MR. PHILLIPS: Thank you, Arthur, and the panelists. I'd just like to thank all of you for your participation in the meeting, and, particularly, I'd like to thank Cathy, Marca, and Bob for their assistance.

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